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Analysis of the St. Louis RAMS (Regional Air Monitoring System) Ambient Particulate Data
Volume I: Final Report

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Santa Monica, CA

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16. ABSTRACT

In this report, a variety of data analysis methods are used to study the 1976 particulate data from the Regional Air Monitoring System (RAMS) in St. Louis. The aerosol data, collected at ten sites, include hi-vol measurements of total suspended particulate mass (TSP), as well as dichotomous sampler measurements of inhalable particulate mass (IP). IP is subdivided into fine particles (less than 2.4 μm in diameter) and coarse particles (between 2.4 and 20 μm in diameter). This study also includes dichotomous sampler data for eight trace elements (S, Si, Al, Ca, Pb, V, Ti, and Fe) and data for 11 meteorological parameters.

The analyses characterize the spatial pattern of particulate matter in-and-near St. Louis; background aerosol concentrations and particulate transport; temporal patterns of particulate concentrations, the dependence of aerosol concentrations on meteorology; and the relationship between hi-vol data and dichotomous data.

Averaged over the RAMS network, IP mass consists of 50% fine particles and 50% coarse particles. Sulfates, secondary aerosols occurring on a large (air basin and synoptic) scale, constitute 53% of fine mass and 29% of IP. Crustal material, mostly consisting of manmade fugitive dust, comprises 83% of coarse mass and 47% of IP. That sulfate is the major source of fine mass and that crustal material dominates coarse mass are the two major themes apparent in the spatial, temporal, and meteorological patterns of the dichotomous data.

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Volume I:
Final Report

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ABSTRACT

In this report, a variety of data analysis methods are used to study the 1976 particulate data from the Regional Air Monitoring System (RAMS) in St. Louis. The aerosol data, collected at ten sites, include Hi-Vol measurements of total suspended particulate mass (TSP) as well as dichotomous sampler measurements of inhalable particulate mass (IP); IP is subdivided into fine particles (less than 2.4 μm in diameter) and coarse particles (between 2.4 and 20 μm in diameter). This study also includes dichotomous sampler data for eight trace elements (S, Si, Al, Ca, Pb, V, Ti, and Fe) and data for eleven meteorological parameters.

The analyses characterize the spatial pattern of particulate matter in-and-near St. Louis; background aerosol concentrations and particulate transport; temporal patterns of particulate concentrations; the dependence of aerosol concentrations on meteorology; and the relationship between Hi-Vol data and dichotomous data. These analyses, as well as chemical element balance calculations, shed considerable light on the sources of particulate matter in St. Louis.

Averaged over the RAMS network, IP mass consists of 50% fine particles and 50% coarse particles. Sulfates, secondary aerosols occurring on a large (air basin and synoptic) scale, constitute 53% of fine mass and 29% of IP. Crustal material, mostly consisting of man-made fugitive dust, comprises 83% of coarse mass and 47% of IP. That sulfate is the major source of fine mass and that crustal material dominates coarse mass are the two major themes apparent in the spatial, temporal, and meteorological patterns of the dichotomous data.

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EXECUTIVE SUMMARY

The Regional Air Monitoring System (RAMS) operated by EPA in St. Louis has supplied a rich data set for studying ambient particulate concentrations in-and-near a major metropolitan area. In order to provide an overview of the data and to illustrate some of the major implications of the data, this report presents a series of statistical analyses conducted with the RAMS particulate measurements. The statistical methods applied range from simple graphical or tabular displays to complex techniques such as decision-tree/pattern-recognition analysis. The investigation addresses not only aerosol mass concentrations, but also the size distribution and chemical composition of the particulate matter.

The data in this report include aerosol measurements for the year 1976 taken at the RAMS sites plus 1976 meteorological data collected at both the RAMS sites and Lambert International Airport. Ten of the RAMS stations monitored particulate matter; based on distance to the center-city, we have classified five of these sites as urban, three as suburban, and two as rural. Each of the RAMS particulate stations was equipped with two aerosol mass monitors: a Hi-Vol sampler measuring total suspended particulate mass (TSP), and a dichotomous sampler measuring inhalable particulate mass (IP). The TSP data essentially represent the mass concentration of particles less than approximately 50 μm in diameter. The IP measurements, in total representing the mass of particles less than approximately 20 μm in diameter,* consist of two parts: mass of fine particles (less than 2.4 μm in size) which we denote as FINE, and mass of coarse particles (between 2.4 and 20 μm in size) which we denote as COARSE. In addition to TSP, IP, COARSE, and FINE, this study analyzes data for eight of the chemical elements measured as part of the dichotomous sampler program. Although some of these elements stem from several source types, we have found that, in St. Louis, these elements serve as key tracers for the following source types: sulfate aerosol (sulfur), shale-type crustal material (silicon and aluminun), limestone-type crustal material (calcium), automotive exhaust (lead), fuel oil combustion (vanadium), paint pigment plant (titanium), and iron/steel industry (iron). As described in a companion report (Eldon et al., 1979), all the particulate data were subjected to a data quality analysis before being used in this study.

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*Actually, our use of the term "inhalable particulate mass" involves a slight misnomer. EPA (Miller et al., 1979) has recently recommended that particles less than 15 μm in diameter be defined as inhalable particulate matter. Although the type of dichotomous sampler used in St. Louis had a 20 μm rather than a 15 μm upper size cut-off, for convenience we will nevertheless refer to these data as IP.

The specific purposes of this study are to investigate the spatial patterns of particulate matter in-and-near St. Louis (Chapter 2); to estimate background aerosol concentrations and examine particulate transport (Chapter 3); to characterize the temporal patterns of particulate concentrations (Chapter 4); to analyze the dependence of particulate concentrations on meteorology (Chapter 5); to determine the relationship between Hi-Vol data and dichotomous data (Chapter 6); and, finally, to assess the contributions of various source types to ambient aerosol concentrations (Chapter 7). The following subsections summarize our findings and conclusions. For convenient referal, the summary is organized according to the order of the chapters, except Chapter 7 (Sources of Particulate Matter in St. Louis) is presented first. We discuss the source assessment first because many of our most important conclusions concern the origins of particulate matter, and because an understanding of sources facilitates the discussions of the other chapters.

Sources of Particulate Matter

Annual mean concentrations of IP range from 29 to 60 $\mu g/m^3$ among the RAMS sites.[*] Averaged over the RAMS network, IP consists of 50% FINE and 50% COARSE. The chemical element balance of Dzubay (1979), modified to include data for the entire year of 1976,[**] implies that three-fourths of IP consists of two components -- sulfate (29%) and crustal material (47%).

---

[*] Note that all averages in this report represent arithmetic means rather than geometric means.

[**] All further allusions to chemical element balance in this summary will refer to Dzubay's results as modified herein to cover the entire year of 1976.

FINE consists basically of man-made particulate matter, especially secondary aerosols (aerosols formed from gas-to-particle conversion), especially sulfates. Chemical element balance calculations imply that, averaged over the year and over all RAMS sites, sulfate constitutes 53% of FINE. That sulfur (or sulfate) is the major determinant of FINE is also strongly evidenced by several other analyses in this report. These analyses further indicate that sulfate is mostly secondary aerosol (formed from SO_2 emissions) rather than primary aerosol (directly emitted), and that sulfate tends to be a large scale phenomenon -- partly air basin in scale and partly synoptic in scale. Because electric power plants contribute approximately 80% of SO_x emissions in the St. Louis AQCR and 70% on the larger synoptic scale, they are likely candidates as the major source of sulfate.

The chemical element balance and other analyses demonstrate that no single type of aerosol dominates the non-sulfate 47% of FINE. Rather, the non-sulfate part of FINE appears to be a mixture of fairly small contributions from numerous sources: secondary organic aerosols, secondary nitrates, automotive exhaust particles, particles from stationary source fuel combustion, the fine fraction (i.e. the lower tail of the size distribution) of suspended dust, etc.

The chemical element balance shows that 83% of COARSE is crustal material, about two-thirds shale-type and one-third limestone-type. Numerous spatial, temporal, and meteorological patterns in the data support the conclusion that COARSE is dominated by crustal material, mostly shale-type. Our analyses further suggest that the crustal material arises primarily from ubiquitous area sources of fugitive dust rather than industrial sources of fly ash, and that most of the fugitive dust is man-made rather than windblown. Despite the omnipresence of automotive traffic, motor vehicles apparently do not constitute a predominant source of fugitive dust. Rather, a variety of dust sources -- paved roads, unpaved roads, quarries, construction, agriculture, soil dust, and certain industries -- seems implicated. A reason why traffic is not the predominant source of dust may be that the RAMS sites (unlike many typical urban particulate monitors) are located away from major roads. Limestone-type dust appears to be more closely associated with commercial/industrial activity (possibly cement manufacturing and construction) than shale-type dust.

The remaining, non-crustal 17% of COARSE stems from a wide variety of sources. The data suggest that most of these sources are man-made.

In the sense that crustal material and sulfates are the two major sources, the origins of IP are generally similar at the urban and rural locations. The relative importance of crustal material and COARSE is slightly greater at urban sites (basically because of more limestone-type dust), while the relative importance of sulfate and FINE is slightly greater at rural sites. There do exist, however, some significant urban/rural differences in particulate matter. Primary aerosols from auto exhaust, fuel combustion, and industrial sources are typically three to five times greater at urban locations. Particulate matter from these sources is not a dominant contributor to IP mass, but it may be of concern because of its chemical nature. The specific sources of fugitive dust also display distinct urban/rural differences; dust in the center-city seems to stem mostly from urban activities, while dust in the countryside likely arises more from agriculture and natural sources (i.e. wind-blown dust).

Our analyses indicate that dust is the dominant source of TSP. Although our analyses show that IP is less affected by fugitive dust (especially wind-blown dust) than TSP, fugitive dust nevertheless contributes significantly to IP (nearly one-half of IP is crustal material, most of which appears to be dust). Air quality standards for IP, therefore, will not completely eliminate the fugitive dust problems associated with the TSP standard.

Spatial Patterns

The one outstanding feature in the spatial patterns for TSP, IP, FINE, and COARSE is a concentric urban bubble; all four particulate variables exhibit nearly monotonic decreases from site 103 (near the center of the RAMS network) radially out to the rural sites. One major reason for this smooth concentricity is that the RAMS sites are generally isolated from strong localized sources (fugitive dust or otherwise). In this sense, the RAMS particulate sites are not fully representative of existing Hi-Vol sites nationwide, many of which are significantly affected by localized sources (especially road dust).

As must be, the spatial trends for IP are intermediate to those for its two components, FINE and COARSE. Because sulfate is the major component of FINE, the spatial pattern of FINE agrees most nearly with that of sulfur; these two variables exhibit the least urban/rural differences. Sulfur at the rural sites averages 75% of sulfur at the urban sites, reflecting the large (air basin and synoptic) geographical scale of sulfate.

The spatial trends of COARSE closely resemble those of its predominant constituent, crustal material (as traced by the elements Si, Al, and Ca). On the average, urban sites are about 70% enriched over the rural sites in crustal-shale and 90% enriched in crustal-limestone. The smooth concentric pattern exhibited by COARSE and the crustal elements suggests that crustal material originates more from widespread area sources of fugitive dust than from concentrated industrial sources of fly ash.

The elements Pb (auto exhaust), V (fuel oil), Ti (paint pigment plant), and Fe (iron/steel industry) all display erratic site-to-site fluctuations and large urban/rural differences (averaging a factor of about three to five). The dissimilarity in the spatial patterns for these elements versus IP, FINE, and COARSE reflects the fact that none of the sources associated with these elements are major contributors to IP.

To determine the effect of monitor site characteristics, the particulate data can be sorted with respect to both location in the metropolitan area (urban vs. suburban vs. rural) and local environment (industrial vs. commercial vs. residential vs. agricultural). For TSP, IP, FINE, COARSE, and the eight elements, this sorting reveals systematic increases from rural to suburban to urban locations but no substantial variation with type of local environment.

The spatial scale of factors affecting particulate concentrations can be investigated through interstation correlations (i.e. correlations of daily particulate variations between pairs of sites). Interstation correlations are extremely high for sulfur and high for FINE, reflecting the large spatial scale of sulfates and the importance of sulfates to FINE. The correlations are moderately high for Si, Al, Ca, COARSE, and TSP, evidently reflecting the common factor (area sources of fugitive dust) affecting all these variables. Lead and iron exhibit moderate interstation correlations, while titanium and vanadium demonstrate moderate to low correlations. One would expect elements with low correlations to be associated with spatially concentrated sources (e.g. point sources).

Background Concentrations and Particulate Transport

For days with predominantly northern and southern wind-flows, regional upwind/downwind analysis is used to create particulate concentration profiles across the St. Louis area. The most salient feature of these profiles is an urban "bubble", with relatively lower particulate concentrations in upwind and downwind rural areas.

The regional upwind/downwind studies and pollution rose analyses for the rural sites indicate that the direct dowmwind transport effect of St. Louis on rural areas is approximately 3 to 5 $\mu g/m^3$ of aerosol, essentially all in the fine size range. Because an individual rural location is not always downwind of the center city, the effect of directly transported urban aerosols on the annual mean at a rural site should be somewhat less, possibly 1 to 3 $\mu g/m^3$. Allowing also for "indirect" transport (from the sloshing of air masses during periods of light or variable winds), we conjecture that the total impact of the urban area on annual mean concentrations at the rural sites is on the order of 2 to 5 $\mu g/m^3$ of (basically fine) particulate matter.

The regional upwind/downwind analysis for sulfur suggests that much (and possibly the majority) of sulfate in St. Louis is synoptic in nature. Furthermore, the pollution rose analysis hints that important synoptic scale factors may be the dense SO_x emissions in the Ohio Valley toward the east and high pressure systems moving up from the Gulf of Mexico (these high pressure systems are known to be conducive to large scale sulfate episodes).

Pollution roses for IP, FINE, COARSE, and most of the elements tend to be fairly isotropic (invariant with wind direction). This is expected because the two major components of IP should display rather weak wind-directional dependencies; sulfates tend to be a large scale phenomenon, and crustal material stems from widespread area sources of fugitive dust. Only the pollution roses for titanium display very strong directional biases; these roses implicate the paint pigment plant as the principal source of Ti.

Background particulate concentrations consist of aerosols from natural sources and from man-made sources exterior to the St. Louis AQCR. Our estimate of background concentrations for St. Louis differs considerably from estimates made by Record et al. (1976). Further analysis is needed to resolve this issue. The best guess, however, based on current information would appear to be approximately 35 $\mu g/m^3$ for TSP and 20 $\mu g/m^3$ for IP.

Temporal Patterns

Particulate concentrations in the St. Louis area tend to be highest during the summer (June through September). The summer peak is strong for FINE, moderately strong for IP, moderate for COARSE, and very weak for TSP. Sulfate (sulfur), the major single component of FINE, displays a pronounced summer maximum similar to that of FINE. Crustal material (as traced by Si, Al, and Ca), the predominant source of COARSE, exhibits a moderate summer maximum similar to that of COARSE.

On a weekly basis, the only salient feature in the aerosol data is a significant weekend-weekday effect. For all twelve particulate variables (TSP, IP, FINE, COARSE, and the eight elements), and for the averages over all three types of site (urban, suburban, and rural), weekend mean concentrations are smaller than weekday mean concentrations. For most of the particulate variables, the weekend-weekday differences are greatest for the urban sites (typically about 20%), less for the suburban sites (typically about 10%), and even less for the rural sites (typically about 5%). This most likely reflects the fact that man-made sources are relatively more important in urban areas than rural areas.

Sulfur and FINE are the only variables that show rural weekend-weekday percentage decreases about as large as the urban decreases. This again emphasizes the large (air basin or synoptic) scale of sulfates and other secondary aerosols that constitute the major part of FINE. That substantial (∿ 15%) weekend-weekday differences do exist for sulfate indicates that a significant part of sulfate is of air basin scale rather than synoptic scale; we would expect synoptic sulfates to display little weekend effect due to a time lag between emission changes and observed air quality changes.

The weekend-weekday differences for COARSE are fairly similar to those of the crustal elements (Si, Al, and Ca). The significant weekend-weekday differences for crustal material suggest that most of the crustal material (at least for urban sites) stems from man-made sources. The weekend-weekday differences are particularly large for Ca, indicating that crustal limestone is more closely associated with commerical and industrial activities than crustal shale is.

Weekend-weekday differences are rather slight for TSP. This may signify that TSP is more affected by wind-blown dust than is COARSE or IP.

The only particulate variable showing a pronounced diurnal pattern is lead which demonstrates a distinct afternoon (noon-6 PM) minimum. There is no obvious reason why lead uniquely exhibits this pattern. That crustal material displays a different diurnal pattern (as well as different spatial patterns) from lead hints that road dust is not the overwhelming source of fugitive dust.

Decision-Tree Analysis of Particulate and Meteorological Data

The CART (Classification and Regression Trees) program developed at Technology Service Corporation is used to conduct decision-tree studies. The CART program offers several advantages over more conventional data-analytic techniques such as multiple linear regression. CART is non-parametric and nonlinear; the decision-trees are easily interpreted on physical grounds; the CART results are evaluated by cross-validation tests with independent data sets; and decision-trees tend to explain more variance than multiple linear regression.

Using the CART program to relate daily concentrations of IP and FINE to daily elemental concentrations yields very good statistical fits; the overall correlation coefficients* are ∿ 0.83 for FINE and ∿ 0.81 for IP in cross-validation tests. As expected, sulfur stands out as, by far, the most important element explaining the variance in FINE. Also as expected, sulfur and the crustal elements (Si, Al, and Ca) account for most of the variance in IP.

The application of CART in relating particulate variables to eleven meteorological variables produces less than excellent statistical fits. Cross-validation tests with independent data sets yield correlation coefficients on the order of 0.55 for FINE, IP, and S; 0.40 for COARSE, Si, Al, Ca, and Fe; and less than 0.30 for TSP, Pb, V, and Ti. Despite the less than excellent levels of percent variance explained, the forms of the decision-trees do provide a reasonable and consistent picture of the type of meteorology associated with high particulate levels.

Two basic themes stand out in the decision-tree classes for both high sulfur and high FINE: prolonged stagnations and high temperatures. These results are consistent with sulfate being the major component of FINE (worst-case meteorology is similar for both variables); with sulfate being mostly secondary aerosol (the worst-case meteorological classes are conducive to secondary aerosol formation); and with sulfate and FINE exhibiting pronounced seasonal patterns (prolonged stagnations and high temperatures are basically summertime conditions). Some aspects of the decision-trees for FINE point toward other man-made aerosols (both primary and secondary) as the sources of the non-sulfate fraction of FINE.

The two meteorological conditions producing high levels of COARSE and the crustal elements (Si, Al, and Ca) are dryness and low wind speeds. This implicates anthropogenic sources of fugitive dust as the major contributors to the crustal material that dominates COARSE. The decision-trees also hint that the small non-crustal component of COARSE is man-made.

The decision-tree results for IP are a mixture of the results for FINE and COARSE. The decision-tree classes for high TSP point toward one major factor -- dryness -- indicating that TSP tends to be dominated by fugitive dust.

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*The square of the correlation coefficient represents the percent of variance explained in the dependent variable.

Relationship Between TSP and IP

On a day-to-day basis, TSP is not a very good predictor of IP, COARSE, and (especially) FINE. Correlations between daily Hi-Vol and dichotomous measurements at individual RAMS sites average 0.67 for TSP versus IP, 0.69 for TSP versus COARSE, and 0.47 for TSP versus FINE.

One way of characterizing the relationship between TSP and IP is to study the properties of the IP/TSP ratio. The annual mean IP/TSP ratio, averaged over all RAMS sites, is 0.61. At individual sites, the day-to-day standard deviation away from the annual mean is ± 0.22 (± 36% of the annual mean). The site-to-site standard deviation of the annual mean ratios, however, is only ± 0.06 (± 10% of the network annual mean). Thus, for the St. Louis RAMS stations, TSP tends to be a fair predictor of IP on an annual mean basis even though it is a poor predictor on a daily basis.

The IP/TSP ratio does not display an obvious geographical pattern within the St. Louis region nor does it show consistent variations according to site characteristics. The IP/TSP ratio also lacks variation with respect to day of the week. The ratio does, however, exhibit a seasonal pattern, being higher in the summer months. The summer maximum in IP/TSP (meaning that a greater fraction of the ambient aerosol is less than 20 μm in the summer) is partly due to the pronounced sulfate peak during the summer.

Decision-tree analyses relating the IP/TSP ratio to eight elemental variables and to eleven meteorological parameters explain very little variance in IP/TSP. The analyses do suggest, however, that days with high IP/TSP ratios are days with high sulfate concentrations.

1.0 INTRODUCTION

The Regional Air Monitoring System (RAMS) operated by EPA in St. Louis has provided a wealth of data regarding ambient particulate matter in-and-near a major metropolitan area. In order to obtain a basic understanding of the data and to see what fundamental implications can be drawn from the data, this report presents a series of statistical analyses conducted with the RAMS particulate measurements. The analyses address not only particulate mass concentrations, but also the size distribution and chemical composition of the particulate matter. The specific purposes are to investigate the spatial and temporal patterns of the aerosol in a major urban area; to determine background particulate concentrations and examine particulate transport relationships; to study the dependence of ambient particulate matter on meteorology; to understand the relationship between Hi-Vol data and dichotomous data; and, finally, to assess the contributions of various source types to ambient aerosol concentrations.

1.1 THE RAMS PARTICULATE DATA

As shown in Figure 1.1, the RAMS air quality and meteorological network consisted of 25 air monitoring stations (numbered 101 through 125). Station 101 resided at the center of the network, and the remainder of the stations formed four approximately concentric circles at radii of 4, 10, 20, and 40 km (Hern and Taterka, 1977). Only ten of the RAMS stations (sites 103, 105, and 106 in the first circle; sites 108 and 112 in the second circle; sites 115, 118, and 120 in the third circle; and sites 122 and 124 in the fourth circle) monitored particulate matter. The aerosol data from these ten stations serves as the subject of this report.

Two techniques, the Hi-Vol sampler and the dichotomous sampler, were used to measure particulate matter at the RAMS stations. The Hi-Vol measurements are referred to as total suspended particulate mass (TSP) and essentially represent the mass of particles less than approximately 50 μm in diameter. The RAMS dichotomous samplers measure the mass of particles less than approximately 2.4 μm in diameter as well as the mass of particles

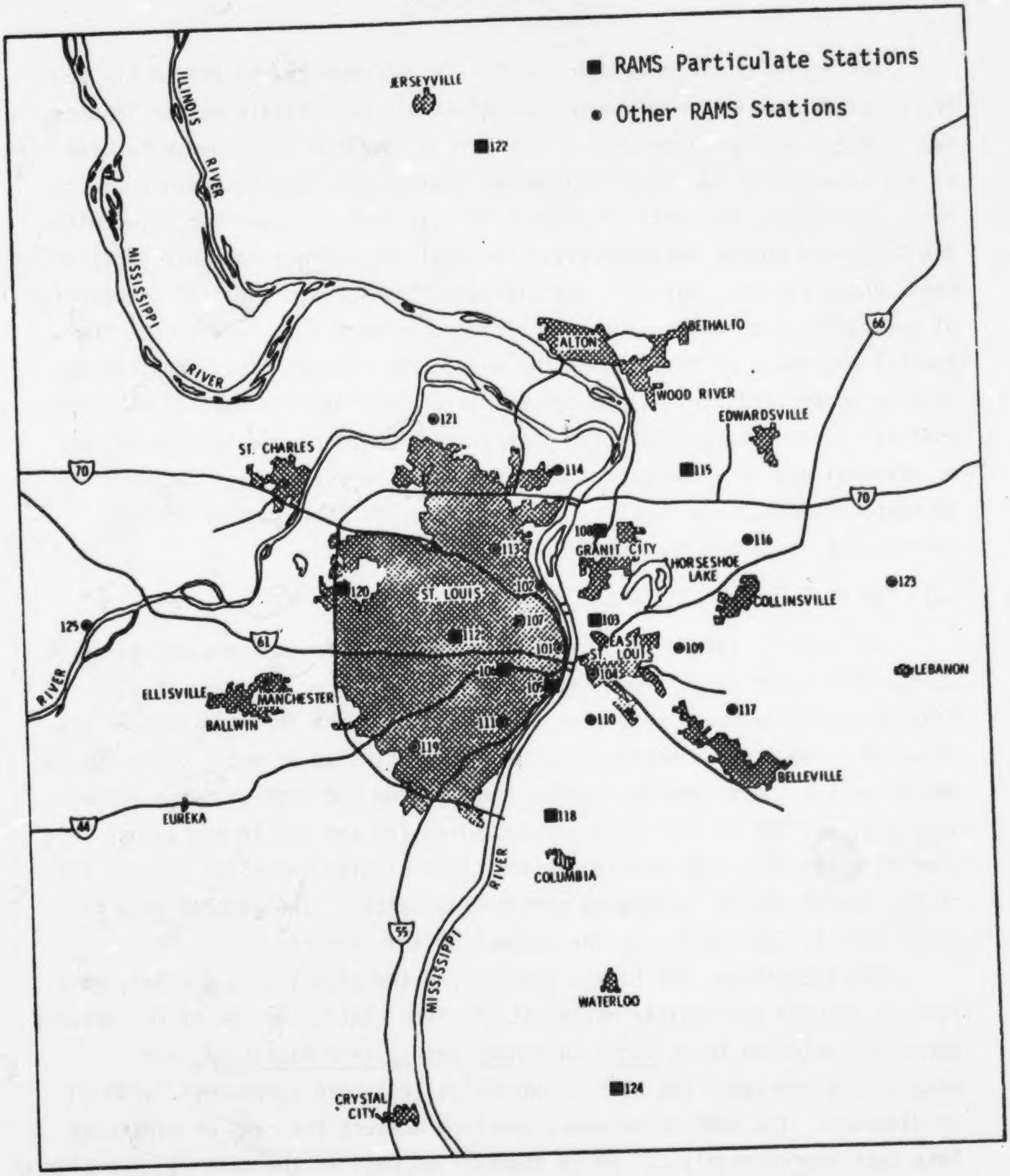
■ RAMS Particulate Stations
● Other RAMS Stations
JERSEYVILLE
122
ILLINOIS RIVER
MISSISSIPPI RIVER
ALTON
BETHALTO
66
WOOD RIVER
EDWARDSVILLE
121
ST. CHARLES
70
114
115
70
108
GRANIT CITY
116
113
HORSESHOE LAKE
ST. LOUIS
102
120
123
COLLINSVILLE
125
103
107
61
112
EAST ST. LOUIS
109
101
LEBANON
RIVER
ELLISVILLE
MANCHESTER
BALLWIN
106
105
104
117
111
110
119
BELLEVILLE
EUREKA
44
118
COLUMBIA
RIVER
MISSISSIPPI
55
WATERLOO
124
CRYSTAL CITY

Figure 1.1 The RAMS monitoring network in St. Louis.

in the approximate size range 2.4 - 20 μm (Dzubay, 1979).[*] Because EPA (Miller et al., 1979) has recently stressed the special importance of inhalable particles ($\lesssim$ 15 μm in diameter) and fine particles ($\lesssim$ 2.5 μm in diameter), and because the extensive set of dichotomous measurements is the most significant novel feature of the RAMS data, this report emphasizes the dichotomous data more than the Hi-Vol data. Even though the RAMS dichotomous samplers had an upper size cut-off of approximately 20 μm rather than 15 μm, we will refer to the total dichotomous mass as inhalable particles (IP). The two size fractions, less than 2.4 μm and 2.4 to 20 μm, will be called fine particle mass (FINE) and coarse particle mass (COARSE), respectively.

The RAMS Hi-Vol samplers were run for 24 hours, every third day. Although the dichotomous samplers essentially measured 12-hour averages on a continuous basis,[**] most of the analyses performed herein use only 24-hour averaged dichotomous data. The RAMS network operated from mid-1975 to early 1977, but this report includes data only for the year 1976.

The RAMS dichotomous data (both the fine and coarse fractions) were analyzed by X-ray spectroscopy to determine the concentrations of 28 elements (Goulding et al., 1978). In this report we will use data for eight of the elements that are especially important in determining the nature of particulate sources. These eight elements, and the source types in St. Louis that the elements are useful in identifying, are listed in Table 1.1.

This study also utilizes data on eleven meteorological parameters. These parameters are defined in Table 1.2.

1.2 DATA QUALITY ANALYSIS

As a preliminary to this study, Eldon et al. (1979) performed a data quality analysis on the 1976 RAMS Hi-Vol and dichotomous data. The data quality analysis consisted of three parts. First, the five highest and

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[*] It should be noted that the upper size cut-off at 20 μm may be sensitive to wind speed; specifically, the collection efficiency of particles near the upper size cut-off may decrease with increasing wind speed.

[**] At two of the stations (103 and 105), 6-hour samples rather than 12-hour samples were collected. Also, during an intensive study period in the summer of 1976, sites 103, 105, and 112 had 2-hour samples while most of the other sites had 6-hour samples.

TABLE 1.1 TRACE ELEMENTS INCLUDED IN THIS STUDY.

ELEMENTS	SOURCES IN ST. LOUIS THAT CAN BE IDENTIFIED BY THE ELEMENTS* (In order of decreasing importance as to usefulness of the element in identifying the source. Underlined sources especially important.)
Sulfur (S)	Sulfate aerosol (basically a secondary aerosol from gaseous SO_x emissions). Note: If one assumes all sulfur is $(NH_4)_2SO_4$, then [sulfate] = 4.1[sulfur]; if one assumes a representative mixture of sulfates, then [sulfate] $\cong$ 3.9[sulfur](White and Roberts, 1977).
Silicon (Si)	Soil dust (shale type), coal combustion.
Aluminum (Al)	Soil dust (shale type), coal combustion.
Calcium (Ca)	Cement dust, soil dust (limestone type), coal combustion.
Lead (Pb)	Auto exhaust.
Vanadium (V)	Fuel oil combustion.
Titanium (Ti)	Paint pigment plant, soil dust, coal combustion.
Iron (Fe)	Iron and steel industry, soil dust, coal combustion.

*In compiling this table, we relied mostly on a paper by Dzubay (1979), but also considered the work of Miller et al. (1972), Friedlander (1973), Gatz (1975), and Kowlaczyk et al. (1978).

TABLE 1.2 METEOROLOGICAL PARAMETERS INCLUDED IN THIS STUDY.

SYMBOL	DESCRIPTION	UNITS	LOCATION
AV WD SP	Average Wind Speed: daily 24-hour vector average surface wind speed	m/sec	Individual RAMS Sites
WIND DIR	Wind Direction: most common direction (8-point compass) from which the hourly average surface wind blew during each day (24-hour period).	no units	Individual RAMS Sites
MIXHT AM	Morning Mixing Height: six AM inversion base height as estimated from noon temperature sounding and hourly ground temperature readings.	meters	Lambert Field
MIXHT PM	Evening Mixing Height: six PM inversion base height as estimated from noon temperature sounding and hourly ground temperature readings.	meters	Lambert Field
MIN TEMP	Minimum Temperature: lowest hourly average surface temperature reading for the day.	°C	Lambert Field
MAX TEMP	Maximum Temperature: highest hourly average surface temperature reading for the day.	°C	Lambert Field
%REL HUM	Relative Humidity: daily (24-hour) average of hourly relative humidity readings.	%	Lambert Field
PRECIPIT	Precipitation: whether or not there was measureable precipitation during the day (0 = no, 1 = yes).	no units	Lambert Field
#DAYS PP	Days Since Previous Precipitation: the number of days since the most recent measurable precipitation, irrespective of rain on day in question (i.e. variable ranges from 1 to ∞).	no units	Lambert Field
D7AM PRS	Pressure Difference: today's 7 AM surface barometric pressure minus yesterday's 7 AM pressure.	millibars	Lambert Field
#DY CALM	Days of Calm: the number of consecutive dalm days (from 0 to ∞). A calm day is one where vector average wind speed is below 3 meters/second.	no units	Lambert Field

five lowest values for each variable (i.e. TSP, FINE, COARSE, fine S, coarse S, fine Si, coarse Si, etc.) were printed out along with several preceeding and following measurements at the same site, and with simultaneous measurements at nearby locations. These values were examined manually for consistency and a few erroneous readings (e.g. zeros that should have been blanks and some inexplicably very high values) were deleted.

Second, scatterplots of IP versus TSP at individual locations were prepared. These scatterplots revealed that 10 to 15 TSP readings were outliers. Also, it was noted that TSP samplers did not run for the full 24 hours on twenty occassions (Nelson, 1979). The lists of extreme outliers and data with short sampling times overlapped considerably; both were eliminated from the data set.

Third, a review was conducted of summary statistics at each site (e.g. annual means, average percent fine, etc.), inter-species correlations (e.g. Al vs. Si, S vs. FINE, etc.) at each site, and inter-site correlations for individual aerosol variables to get an overall impression of data quality. This review indicated that the data were generally of good quality despite the existence of the few erroneous values noted earlier.

1.3 SUMMARY OF ANNUAL MEANS

Before proceeding with the statistical analyses of subsequent chapters, it is wothwhile to get a feel for the magnitude of the aerosol variables by examining summary statistics. Table 1.3 lists annual means* for the twelve aerosol variables considered in this study. Appendix A presents more detailed summaries of annual and monthly statistics for both the particulate and meteorological variables.

As indicated in Table 1.3, annual mean TSP concentrations range from 53 $\mu g/m^3$ to 96 $\mu g/m^3$ and average 79 $\mu g/m^3$ among the sites. To ascertain the attainment status of the sites with respect to the present National Ambient Air Quality Standards (NAAQS) for TSP, one must consider annual geometric means and second highest daily maximum concentrations. Data presented in Appendix A indicate that the primary NAAQS for the annual geometric

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*Note that arithmetic means rather than geometric means are used throughout this report.

TABLE 1.3 ANNUAL ARITHMETIC MEAN PARTICULATE CONCENTRATIONS.

ANNUAL MEAN CONCENTRATIONS ($\mu g/m^3$) FOR 1976*

	Site 103	Site 105	Site 106	Site 108	Site 112	Site 115	Site 118	Site 120	Site 122	Site 124	AVERAGE ALL SITES
TSP	96	88	77	80	79	60	66	53	55	53	79
IP	60 (47%)	46 (50%)	48 (50%)	48 (50%)	44 (49%)	38 (52%)	35 (51%)	37 (55%)	33 (51%)	29 (56%)	42 (50%)
FINE	28	23	24	24	21	19	18	20	17	16	21
COARSE	32	23	25	24	23	19	17	17	17	13	21
Sulfur	3.5 (83%)	3.1 (88%)	3.5 (88%)	3.3 (89%)	2.9 (89%)	2.8 (92%)	2.7 (91%)	2.7 (91%)	2.5 (92%)	2.4 (93%)	2.9 (90%)
Silicon	4.8 (7%)	3.6 (8%)	4.2 (10%)	4.1 (8%)	4.3 (8%)	3.5 (9%)	2.9 (9%)	2.7 (8%)	3.0 (7%)	2.2 (20%)	3.5 (9%)
Aluminum	1.5 (12%)	1.1 (16%)	1.1 (14%)	1.2 (13%)	1.3 (16%)	0.9 (14%)	0.8 (14%)	0.8 (17%)	0.8 (11%)	0.6 (22%)	1.0 (15%)
Calcium	3.3 (5%)	2.7 (5%)	2.5 (5%)	2.9 (5%)	2.0 (6%)	1.7 (8%)	2.1 (9%)	1.5 (6%)	1.9 (6%)	1.0 (24%)	2.2 (8%)
Lead	.68 (79%)	.81 (78%)	.87 (76%)	.75 (80%)	.89 (79%)	.40 (83%)	.37 (79%)	.65 (81%)	.21 (86%)	.19 (82%)	.58 (80%)
Vanadium	.007 (46%)	.006 (60%)	.006 (58%)	.010 (51%)	.007 (55%)	.005 (59%)	.003 (54%)	.003 (58%)	.002 (57%)	.002 (52%)	.005 (55%)
Titanium	.16 (14%)	.21 (16%)	.53 (15%)	.13 (16%)	.23 (16%)	.09 (17%)	.17 (17%)	.09 (16%)	.06 (14%)	.04 (15%)	.17 (16%)
Iron	1.8 (13%)	1.2 (19%)	1.4 (20%)	1.7 (17%)	1.1 (17%)	0.8 (19%)	0.7 (19%)	0.6 (17%)	0.6 (18%)	0.4 (31%)	1.0 (19%)

*Numbers in parentheses represent percent of the dichotomous concentration that is fine.

mean (75 $\mu g/m^3$) was exceeded in 1976 at site 103 (89 $\mu g/m^3$) and site 105 (82 $\mu g/m^3$). The primary NAAQS for daily maximal concentrations (260 $\mu g/m^3$) was not exceeded at any of the RAMS sites in 1976.

The concentrations of IP average 42 $\mu g/m^3$ among the RAMS sites. Approximately half of IP (slightly less than half at center-city sites, slightly more than half at rural sites) consists of fine particles. Sulfur and lead occur predominantly among the fine particles. Silicon, aluminum, calcium, titanium, and iron are found predominantly among the coarse particles. Vanadium occurs nearly equally in COARSE and FINE.

1.4 CHEMICAL ELEMENT BALANCE

One of the objectives of this study is to perform a qualitative assessment of the sources contributing to ambient particulate matter based on the spatial/temporal patterns in aerosol mass and composition, the dependence of aerosol mass and composition on meteorology, particulate transport relationships, etc. A more direct and quantitative empirical technique for assessing source contributions is the chemical element balance (tracer element) method (Miller et al., 1972; Friedlander, 1973; Gatz, 1975; Kowalczyk, 1978). Work on the St. Louis RAMS data with the chemical element balance method is being carried out separately by other researchers (Dzubay, 1979; Gordon, 1979; Hopke, 1979). Although chemical element balance is not within the planned scope of this study, a brief review of preliminary conclusions reached by the tracer element technique is useful for placing our results in better perspective.

Dzubay (1979) performed a tracer element analysis with the RAMS dichotomous data for July and August 1976. He included six types of sources in his analysis: ammonium sulfate, crustal shale (soil, road dust, quarry dust, fly ash, etc.), crustal limestone (calcium-rich soil, cement manufacturing, vehicle generated cement dust, etc.), motor vehicle exhaust, iron/steel industry, and paint pigment. The results are summarized in Figure 1.2. The most important sources of IP were found to be ammonium sulfate, which constituted 59% of FINE and 35% of IP, and the crustal components (mostly shale-type), which constituted 83% of COARSE and 43% of IP.

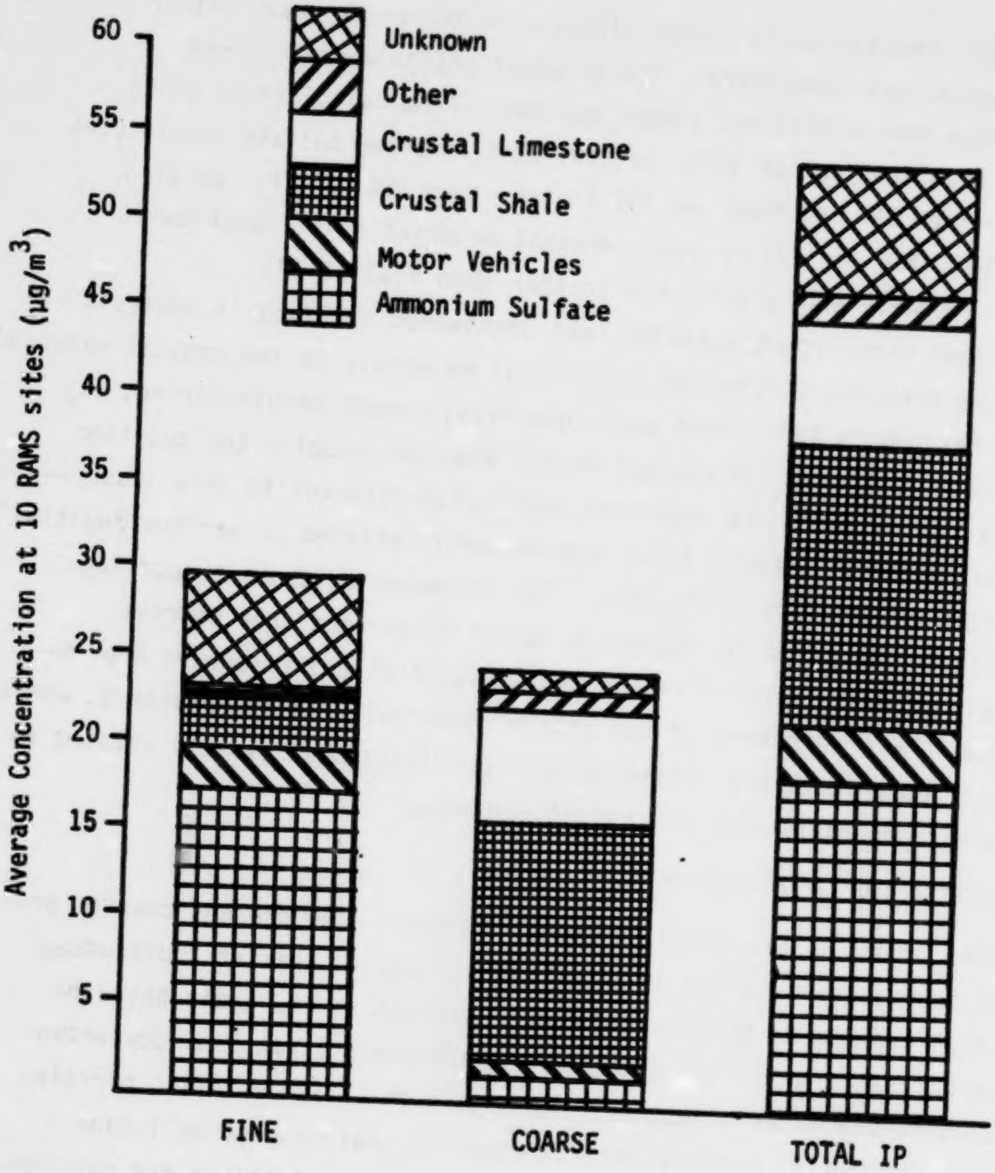

Figure 1.2 Source contributions averaged over the ten RAMS sites for July and August 1976, as determined by chemical element balance (Dzubay, 1979).

Dzubay's results would change slightly if the whole year, rather than July and August were considered. The greatest change would involve sulfates which show a distinct summer maximum. From calculations using data for the entire year of 1976, we find that ammonium sulfate constitutes 53% (rather than 59%) of FINE and 29% (rather than 35%) of IP. We also estimate that, for the entire year, crustal material still constitutes about 83% of COARSE but around 47% (rather than 43%) of IP.

The most significant question left unanswered by Dzubay's analysis concerns the specific origins of the crustal material: Is the crustal material basically related to soil, road dust, quarries, cement manufacturing, fly ash, or what? Although the present report does not resolve the question completely, we do reach one important conclusion relevant to this issue -- the crustal material appears to be predominantly related to various fugitive dust sources rather than to fly ash. The strongest evidence supporting this conclusion is found in Chapter 2, where ubiquitous area sources (e.g. fugitive dust sources) rather than industrial point sources are implicated as the originators of the crustal material, and in Chapter 5, where dryness is found to be the meteorological condition most closely related to high ambient concentrations of crustal elements.

1.5 ORGANIZATION OF THE REPORT

This report is organized in seven chapters. The present chapter provides an introduction to the RAMS particulate data. Chapter 2 discusses the spatial aspects of particulate concentrations in St. Louis based on geographical patterns in the data, the relationship of aerosol concentrations to site characteristics, and the magnitude of interstation correlations. In Chapter 3, regional upwind/downwind analyses and pollution roses are used to study background particulate concentrations and particulate transport. Chapter 4 examines temporal patterns -- seasonal, weekly, and diurnal. Chapter 5 presents the results of pattern-recognition/decision-tree analyses relating aerosol concentrations to meteorological and elemental variables. In Chapter 6, the relationship between Hi-Vol and dichotomous sampler data is studied. Finally, Chapter 7 draws from several parts of the report to synthesize and summarize conclusions regarding the sources of ambient particulate matter in St. Louis.

2.0 SPATIAL PATTERNS OF ST. LOUIS PARTICULATE DATA

The dichotomous and Hi/Vol data from the RAMS network provide a good opportunity to investigate the spatial patterns of ambient particulate matter in-and-near a large metropolitan area. Specific issues that can be addressed include the geographical distribution of ambient particulate matter, the variations among site types, and the spatial scale of aerosol sources.

2.1 GEOGRAPHICAL DISTRIBUTION OF AMBIENT PARTICULATE MATTER

Figures 2.1 through 2.3 illustrate the geographical variation of TSP, IP, and FINE data, respectively.* Concentrations of all three parameters are highest at station 103 and decrease in a fairly concentric manner out to the rural stations (122 and 124).

Because the aerosol spatial distribution appears to be concentric, it is instructive to plot the data versus distance from station 103.** Such plots are presented in Figure 2.4 for all 12 particulate variables considered in this study. To facilitate reading and interpreting the graphs, the variables are divided into three groups: TSP, IP, COARSE, and FINE; S, Si, Al, and Ca; and Pb, V, Ti, and Fe. All the plots are normalized with respect to the concentration at station 124 (the site experiencing the lowest average concentration for all variables except TSP).

Figure 2.4 reveals that TSP, IP, COARSE, and FINE all increase rather monotonically from the rural stations to station 103. This implies that the spatial pattern of the data is indeed very concentric about station 103. One major reason for the smooth concentric pattern is that the RAMS sites tend to be isolated from strong localized sources such as heavily used roads, other intense fugitive dust sources, or industrial point sources (PEDCO, 1978). With the influence of local sources minimized, the gradual increase in ambient particulate matter from rural areas to the center city becomes a dominant spatial feature in the data.

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*Similar maps for eight particulate elements (S, Si, Al, Ca, Pb, V, Ti, and Fe) are presented in Appendix B.

**We also tried plotting the data versus distance from station 101, the center of the entire RAMS network. The plots based on distance to station 101 were not quite as smooth or monotonic as those based on distance to station 103; this apparently indicates that the center of the St. Louis particulate bulge is closer to station 103 than it is to station 101.

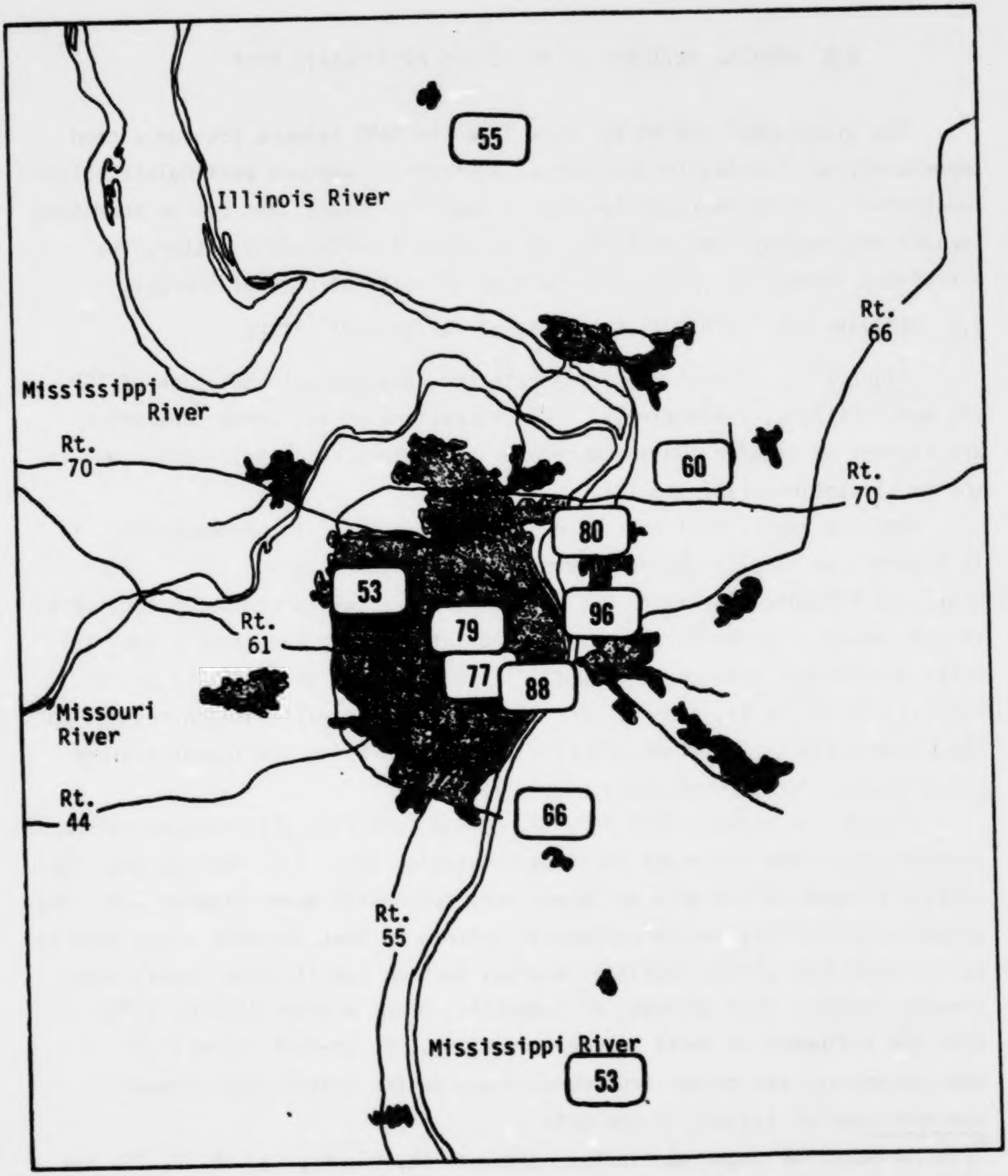
55
Illinois River
Mississippi
River
Rt.
70
Rt.
66
60
Rt.
70
80
53
96
79
Rt.
61
77
88
Missouri
River
Rt.
44
66
Rt.
55
Mississippi River
53

Figure 2.1 Geographical distribution of arithmetic mean TSP mass concentration in the St. Louis area ($\mu g/m^3$).

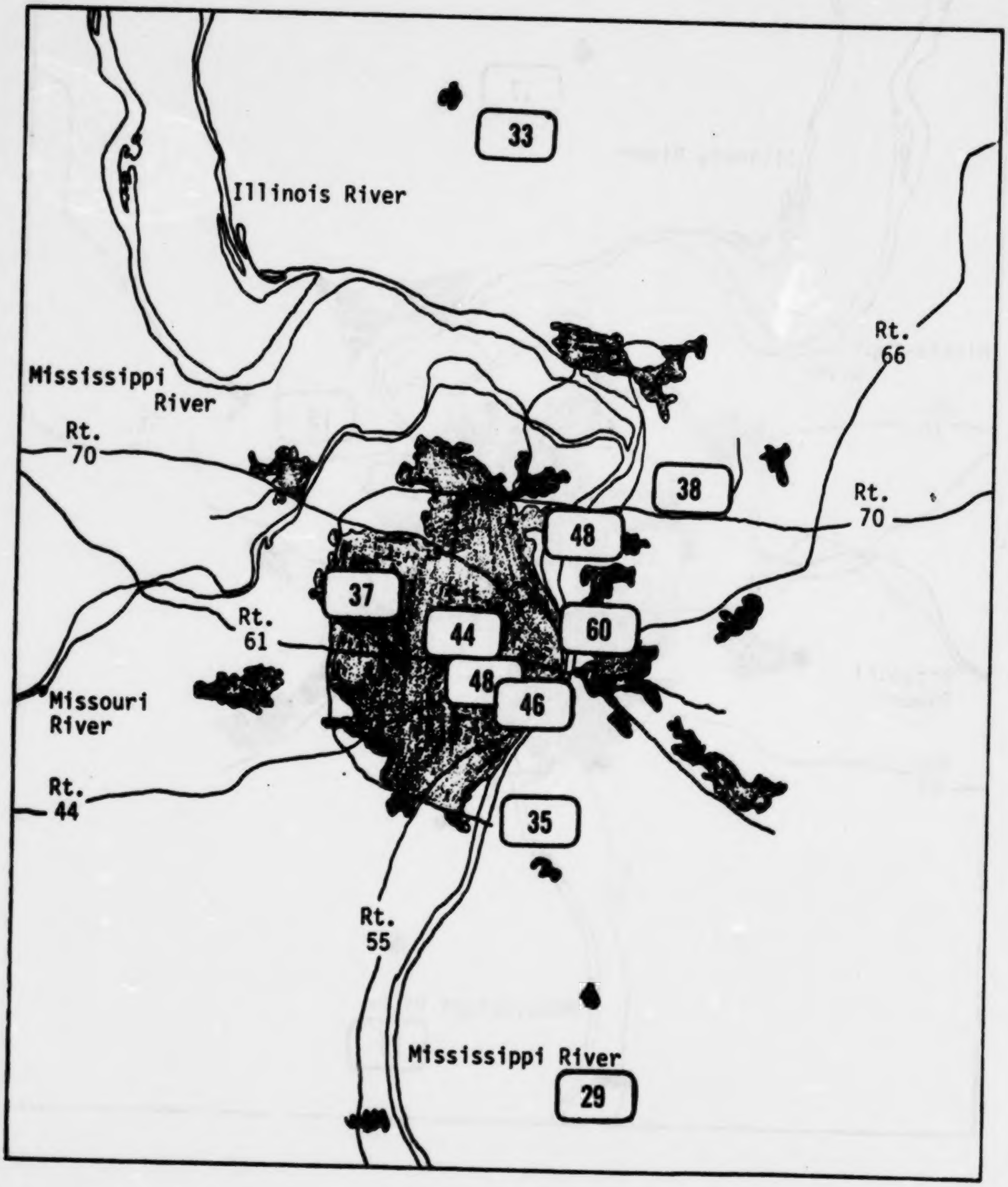
33
Illinois River
Mississippi
River
Rt.
70
Rt.
66
38
Rt.
70
48
37
Rt.
61
44
60
48
46
Missouri
River
Rt.
44
35
Rt.
55
Mississippi River
29

Figure 2.2 Geographical distribution of arithmetic mean IP mass concentration in the St. Louis area ($\mu g/m^3$).

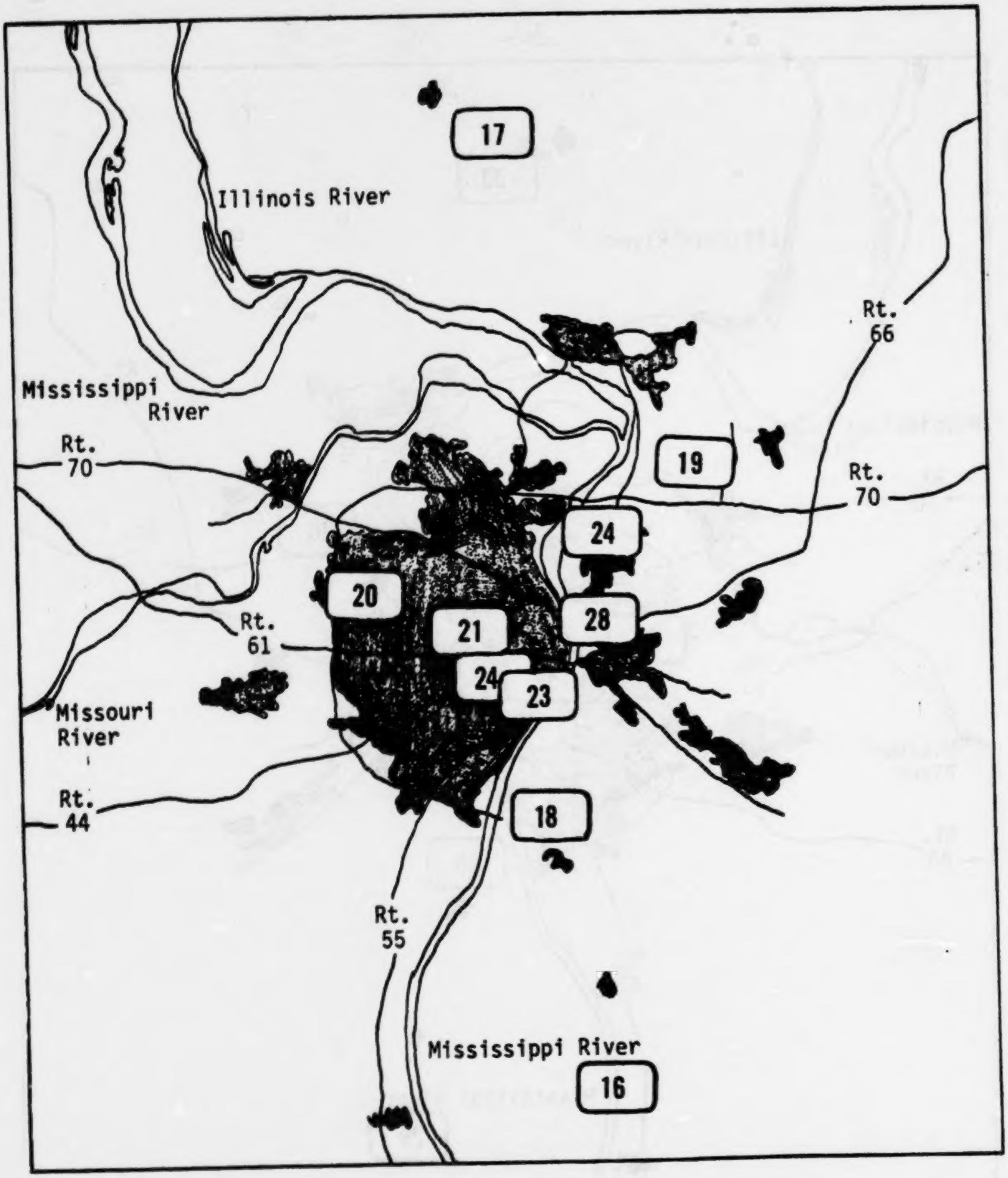
17
Illinois River
Mississippi River
Rt. 66
Rt. 70
19
Rt. 70
24
20
Rt. 61
21
28
24
23
Missouri River
Rt. 44
18
Rt. 55
Mississippi River
16

Figure 2.3 Geographical distribution of arithmetic mean FINE mass concentration in the St. Louis area (μg/m^3).

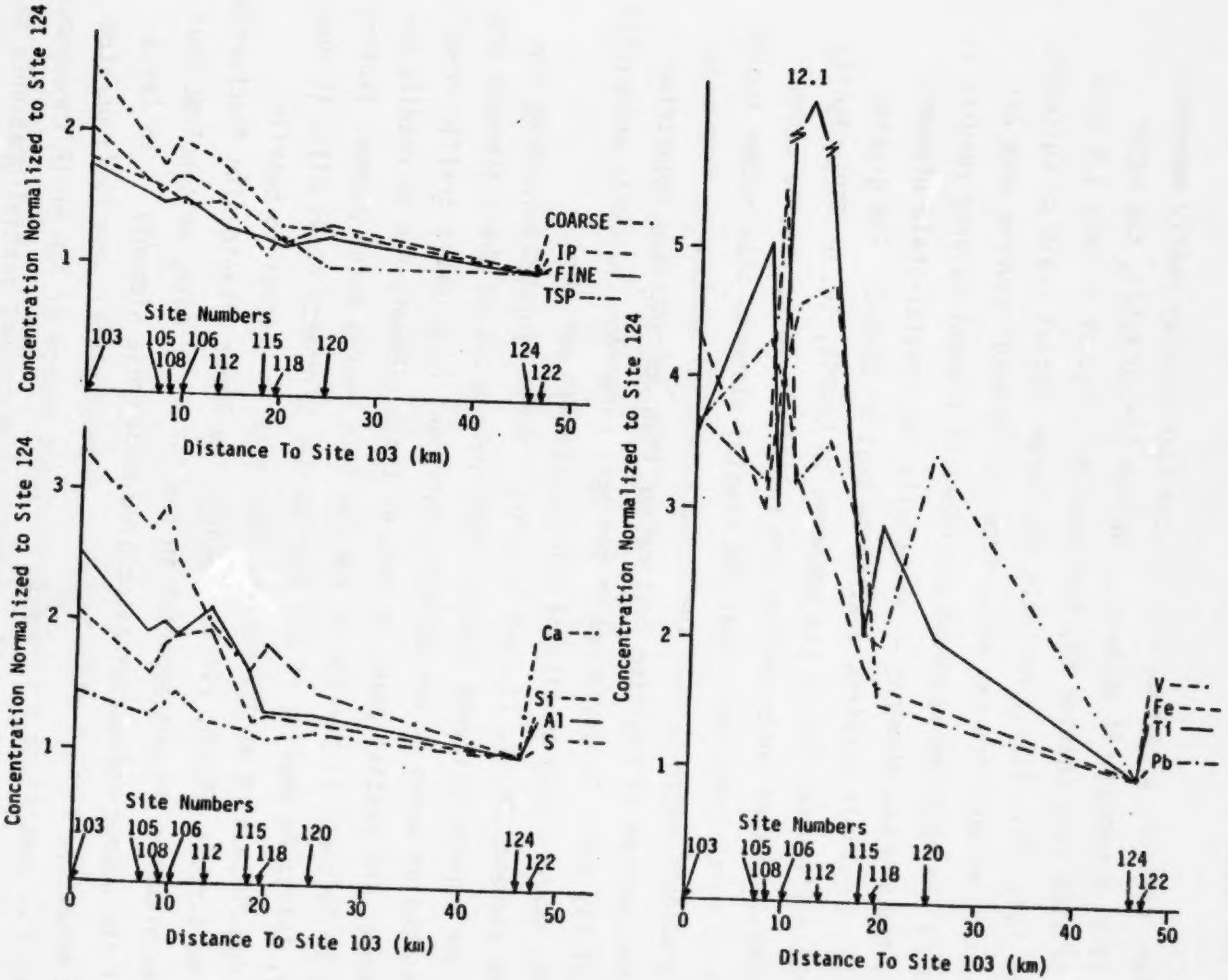

Figure 2.4 Plots of normalized particulate concentrations (with respect to station 124) versus distance from station 103.

The increase in TSP from station 124 to station 103 is by a factor of 1.8. The factor for IP (2.1) is slightly larger and is, of course, intermediate to the factors for COARSE (2.5) and FINE (1.8) which together comprise IP.

Sulfur, silicon, aluminum, and calcium also display nearly monotonic increases (i.e. a concentric pattern). Sulfur (or sulfate), the major component of FINE (see Chapter 1), increases by a factor of only 1.5 from site 124 to 103. This likely reflects the large spatial scale of sulfates; a major portion of sulfates can arise from "background" sources such as power plants. As expected based on the chemical element balance results in Chapter 1, the crustal elements -- especially the crustal-shale elements (Si and Al) -- exhibit patterns similar to that of COARSE. The greater urban/rural differences for Ca (as compared to COARSE, IP, or TSP) signify that crustal-limestone contributions are relatively larger in the center city than at rural locations. That the crustal elements show rather smooth spatial gradients suggests that the crustal material stems more from widespread area sources of fugitive dust rather than concentrated industrial sources of fly ash. As discussed in the next paragraph, elements associated with point sources tend to display erratic spatial patterns.

The elements Pb, V, Ti, and Fe exhibit large fluctuations among the sites. The erratic patterns suggest that the sources of these elements are point sources or areas of concentrated emissions (e.g. dense traffic areas). For example, the spatial peaks in some of these elements can be readily explained as follows: Ti peaking at 106 due to a nearby paint pigment factory (Dzubay, 1979); Fe peaking at 103 and 108 due to nearby steel mills (Dzubay, 1979); and Pb peaking at 112 because that site is closest to heavily travelled streets (PEDCO, 1978). Because the large site-to-site fluctuations of these elements are not apparent in the TSP or IP data, we conclude that none of the source categories associated with these elements -- Pb (automobile exhaust), V (fuel oil), Ti (paint pigment plant), and Fe (iron/steel industry) -- constitute a single predominant source of TSP or IP throughout St. Louis. Rather, TSP and IP evidently show gradual spatial gradients because one large fraction of particulate mass stems from ubiquitous sources of fugitive dust, and because another significant fraction consists of sulfates and other secondary aerosols (which tend to exhibit slight spatial gradients due to the mixing and transport that occurs in the time required for secondary aerosol formation).

2.2 EFFECTS OF SITE CHARACTERISTICS

It is of interest to determine if the spatial variations in ambient particulate matter can be explained, in part, by site characteristics. In examining this question, we will investigate two types of site characteristics: location with respect to the metropolitan area (e.g. urban vs. suburban vs. rural) and local environment (e.g. industrial vs. commercial vs. residential vs. agricultural). We have chosen to classify locations within the first two rings of RAMS stations (sites 103, 105, 106, 108, and 112) as urban, locations in the third ring of RAMS stations (sites 115, 118, and 120) as suburban, and locations in the outer ring of RAMS stations (sites 122 and 124) as rural.* The classification as to local environment is based on detailed site descriptions compiled by PEDCO (1978).

Table 2.1a indicates the distribution of the RAMS particulate stations with respect to site characteristics. Because of the extremely small number of stations involved, there is no hope of obtaining a statistically definitive statement of how particulate levels vary among site types; in fact, half of the site characteristic matrix is barren of stations. Rather, we will attempt only to describe the main qualitative features in the data sorted according to site characteristics.

Tables 2.1b through 2.1d summarize annual mean concentrations of TSP, IP, and FINE, averaged for all sites of each type. Two main tendencies appear in these tables: (1) particulate concentrations increase systematically from rural to suburban to urban locations, and (2) particulate concentrations do not seem to vary substantially with type of local environment. This result reemphasizes a conclusion of the previous section; specifically, we again find that the principal spatial feature of the RAMS data is the concentric decrease in concentrations from urban to rural locations.

We have also examined how the concentrations of the eight trace elements vary with site type (see Appendix C). For all eight elements, we find the same two tendencies apparent in Table 2.1 (i.e. systematic increases from rural to

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*The reader is referred to Figure 1.1 for a map of the RAMS network.

TABLE 2.1 AVERAGE PARTICULATE CONCENTRATION AS A FUNCTION OF SITE TYPE.

Table 2.1a Number of Sites

	Industrial	Commercial	Residential	Agricultural	All
Urban	0	1	2	2	5
Suburban	1	0	0	2	3
Rural	0	0	0	2	2

Table 2.1b Annual Arithmetic Mean TSP ($\mu g/m^3$).

	Industrial	Commercial	Residential	Agricultural	All
Urban	---	88	78	88	84
Suburban	53	---	---	63	59
Rural	---	---	---	54	54

Table 2.1c Annual Arithmetic Mean IP ($\mu g/m^3$).

	Industrial	Commercial	Residential	Agricultural	All
Urban	---	46	46	54	49
Suburban	37	---	---	36	37
Rural	---	---	---	31	31

Table 2.1d Annual Arithmetic Mean FINE ($\mu g/m^3$).

	Industrial	Commercial	Residential	Agricultural	All
Urban	---	23	22	26	24
Suburban	20	---	---	19	19
Rural	---	---	---	16	16

suburban to urban locations, and lack of substantial variations with respect to local site environment).

Table 2.2 summarizes the urban/suburban/rural differences for all twelve variables. Many of the data features noted in the previous section are again apparent in Table 2.2. Specifically, sulfur shows the least spatial variation; FINE shows the second least spatial variation; patterns are similar for COARSE, Si, Al, and Ca; and Pb, V, Ti, and Fe show the greatest urban/rural differences.

2.3 SPATIAL SCALE OF PARTICULATE PHENOMENA

The spatial scale of factors affecting particulate concentrations can be investigated through interstation correlations (i.e. correlations of day-to-day particulate variations between pairs of sites). Table 2.3 summarizes the results of three types of correlation/regression analysis applied to the 24-hour particulate data: (1) the average correlation between each pair of RAMS sites, (2) the average correlation obtained in a multiple regression of each site against its three nearest neighbors, and (3) the average correlation obtained in a multiple regression of each site against its six nearest neighbors. The particulate parameters in Table 2.3 are ordered according to the magnitude of interstation correlations.

As evidenced by Table 2.3, sulfur exhibits extremely high interstation correlations (i.e. all sites exhibit very similar day-to-day fluctuations). This is expected because sulfur basically represents sulfate, a secondary aerosol that tends to show spatially uniform fluctuations on an air basin scale and sometimes even on a synoptic scale. FINE, which consists of approximately 53% sulfate (see Chapter 1), also demonstrates high interstation correlations, although not as high as sulfate (i.e. sulfur). Evidently, the other components of FINE -- other secondary aerosols and primary fine particles -- involve more localized phenomena than does sulfate, leading to lower interstation correlations.

As expected, IP displays a level of correlation intermediate to FINE and COARSE. The variables Si, Al, Ca, COARSE, and TSP all show about the

TABLE 2.2 RURAL VS. SUBURBAN VS. URBAN CONCENTRATIONS OF ST. LOUIS PARTICULATE DATA.*

	TSP	IP	COARSE	FINE
Urban	84 (1.6)	49 (1.6)	25 (1.7)	24 (1.5)
Suburban	59 (1.1)	37 (1.2)	18 (1.2)	19 (1.2)
Rural	54	31	15	16
	S	Si	Al	Ca
Urban	3.3 (1.3)	4.2 (1.6)	1.26 (1.8)	2.7 (1.9)
Suburban	2.7 (1.1)	3.0 (1.2)	0.85 (1.2)	1.7 (1.2)
Rural	2.5	2.6	0.69	1.4
	Pb	V	Ti	Fe
Urban	0.80 (4.0)	0.007 (3.5)	0.26 (5.2)	1.44 (2.7)
Suburban	0.47 (2.4)	0.004 (2.0)	0.12 (2.4)	0.69 (1.3)
Rural	0.20	0.002	0.05	0.53

*First number is concentration in (μg/m^3); second number is ratio to rural.

TABLE 2.3 INTERCORRELATIONS FOR ST. LOUIS PARTICULATE DATA

	AVERAGE CORRELATION COEFFICIENTS		
PARAMETER	Univariate Regression of Each Site Versus All Others	Multiple Regression of Each Site Versus Its Three Nearest Neighbors	Multiple Regression of Each Site Versus Its Six Nearest Neighbors
Sulfur	0.90	0.95	0.96
FINE	0.81	0.90	0.91
IP	0.71	0.86	0.90
Silicon	0.68	0.81	0.83
Aluminum	0.61	0.77	0.80
Calcium	0.56	0.77	0.80
COARSE	0.56	0.77	0.80
TSP	0.58	0.71	0.75
Lead	0.40	0.70	0.76
Iron	0.44	0.64	0.74
Titanium	0.24	0.53	0.65
Vanadium	0.19	0.36	0.47

same, moderately high levels of interstation correlation. Because Si, Al, and Ca are basic tracers for crustal material, this result supports indications elsewhere in this report that the majority of COARSE and TSP consists of crustal material. That the interstation correlations for the crustal elements are much lower that those for sulfates implies that the crustal phenomena are more localized that sulfate phenomena; that these correlations are much higher than for industrial or point source tracers (e.g. V, Ti, and Fe) suggests that the crustal material arises more from area sources of fugitive dust than from industrial or point sources of fly ash.

Lead and iron exhibit moderate interstation correlations, while titanium and vanadium demonstrate moderate to low interstation correlations. We expect elements with low correlations to be associated with spatially concentrated sources (e.g. point sources). Point sources lead to low interstation correlations because there is less tendency for simultaneous high concentrations at all stations (wind direction with respect to the point sources, in addition to overall dispersive conditions, becomes a key meteorological condition).

The larger scale nature of FINE relative to IP, and of IP relative to TSP, may have significant implications with respect to particulate monitoring. Specifically, the scale of representativeness of particulate monitors should be greatest for FINE, less for IP, and least for TSP. This implies that, in characterizing the spatial and temporal patterns of air quality, FINE requires fewer monitors than IP which in turn requires fewer monitors than TSP.

3.0 ANALYSIS OF BACKGROUND CONCENTRATIONS AND PARTICULATE TRANSPORT

The relationship between particulate concentrations and wind direction can provide information about background aerosol concentrations for St. Louis and about particulate transport phenomena that occur in-and-near St. Louis. The wind direction dependence of aerosol concentrations can also identify significant individual sources of particulate matter. This chapter uses two techniques to analyze the relationship between particulate levels and wind direction: (1) regional upwind/downwind analysis and (2) wind rose and pollution rose analysis.

Before starting the technical discussion of this chapter, it is worthwhile to digress a moment and discuss the definition of "background" concentration. For our purposes, background aerosol concentration refers to the aerosol concentration that would exist if all man-made sources in the St. Louis AQCR* were eliminated; background therefore includes contributions from natural sources as well as from man-made sources outside the St. Louis AQCR. Background concentrations tend to be greater than "natural background" but less than concentrations measured at rural locations near St. Louis (because rural aerosol concentrations include particulate matter generated by local man-made sources as well as particulate matter transported from the urban area).

3.1 REGIONAL UPWIND/DOWNWIND ANALYSIS

Regional upwind/downwind analysis is a useful technique for identifying particulate sources, estimating background concentrations, and assessing particulate transport from an urban area. Given a linear set of monitoring sites and data corresponding to wind flow parallel to the line of sites, large sources might be located by noting abrupt concentration increases from one site to the next. Upwind of the source, the concentration naturally should be lower than immediately downwind of it. Upwind/downwind analysis can also help estimate regional background levels of aerosols, since the readings at a rural site, if taken when that site is upwind of the city, should be near the background level. Particulate transport from the urban area can be estimated by the difference between downwind rural concentrations and upwind rural concentrations.

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*Air Quality Control Region

Of the ten RAMS sites equipped with aerosol samplers, six lie approximately in a straight, north-south line through the center of the city. These six sites are 122 (the northernmost rural site), 108, 103, 105, 118, and 124 (the southernmost rural site). Data for these six sites, under predominantly northern and southern flows, serves as the subject of this section. The methodology for calculating concentration cross-sections under northerly and southerly flows is explained in Appendix E.

Figures 3.1 through 3.4 are concentration cross-sections for TSP, IP, FINE, and COARSE. Each figure contains two graphs, one the averages for wind predominantly from the north, the other the averages for wind predominantly from the south. Average concentrations at the central urban site (103), the upwind site (122 for north, 124 for south), and the downwind site (124 for north, 122 for south) are presented in Table 3.1 along with downwind-upwind differences and urban-upwind differences.

The main feature in the cross-sections is the excess of the urban concentrations over the rural concentrations. This feature is demonstrated by the urban "bubble" effect evident in Figures 3.1 through 3.4 and by the significant urban-upwind differences in Table 3.1. The downwind-upwind differences are comparatively minor, indicating that particulate transport from the urban area to rural areas is not an extremely important phenomenon. Although it is difficult to reach definitive conclusions because we are dealing with small differences between large numbers (and therefore the errors tend to be large), the downwind-upwind differences in Table 3.1 generally indicate that the direct downwind transport effect of St Louis on rural areas is approximately 5 $\mu g/m^3$ of aerosol, predominantly in the fine size range.

Because an individual rural location is not always downwind of the center city, the effect of directly transported urban aerosols on the annual mean at a rural site should be less than 5 $\mu g/m^3$, possibly on the order of

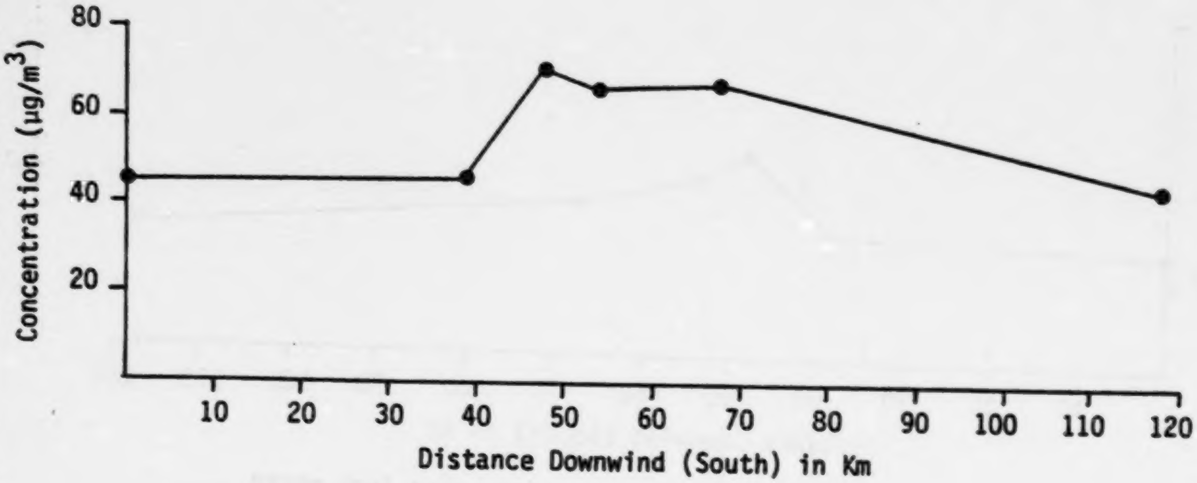

Figure 3.1a TSP concentration cross-section, wind from north.

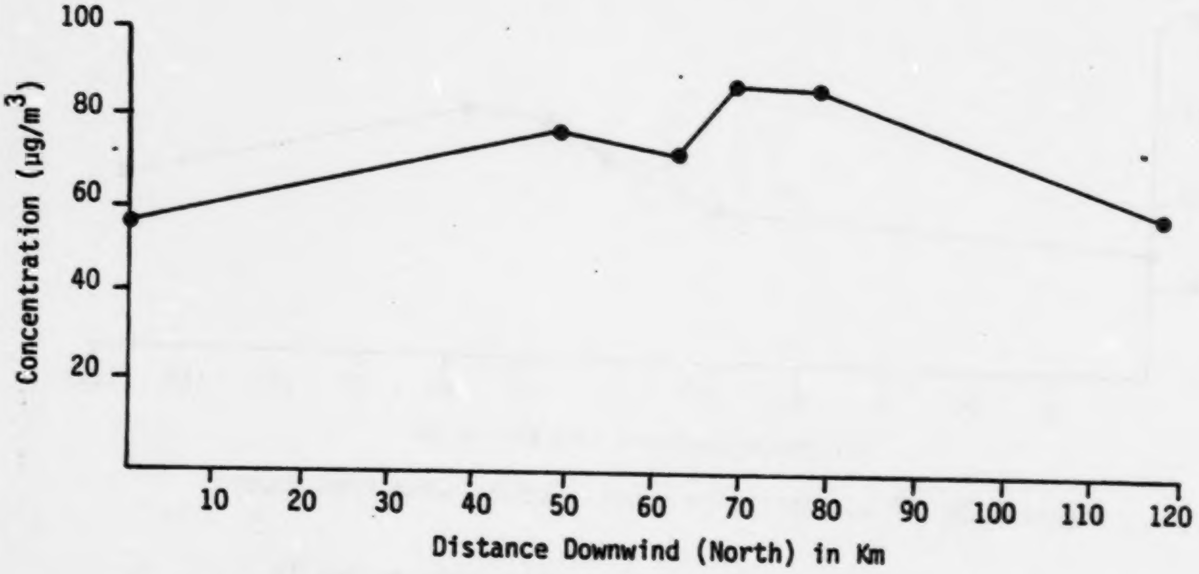

Figure 3.1b TSP concentration cross-section, wind from south.

Figure 3.1 Regional upwind/downwind analyses for TSP.

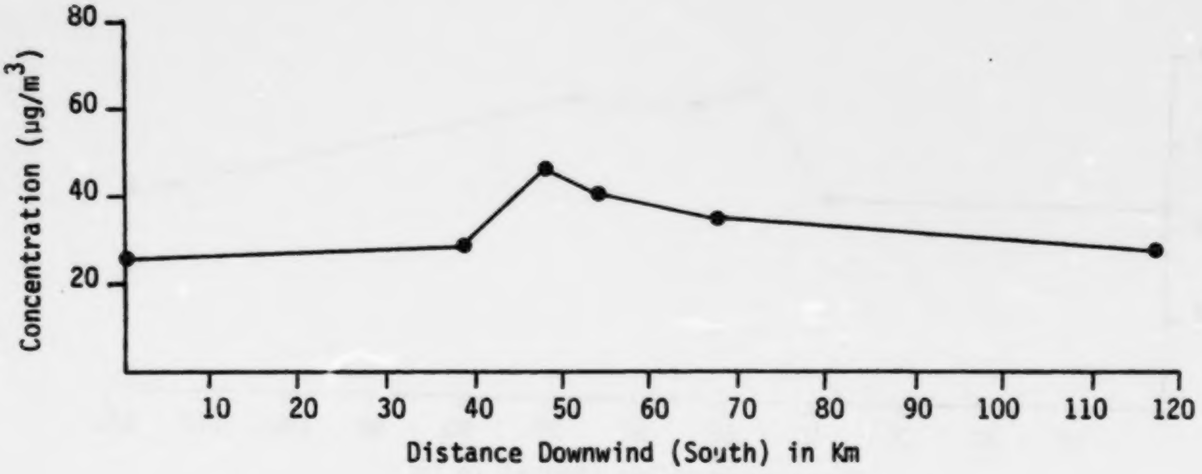

Figure 3.2a IP concentration cross-section, wind from north.

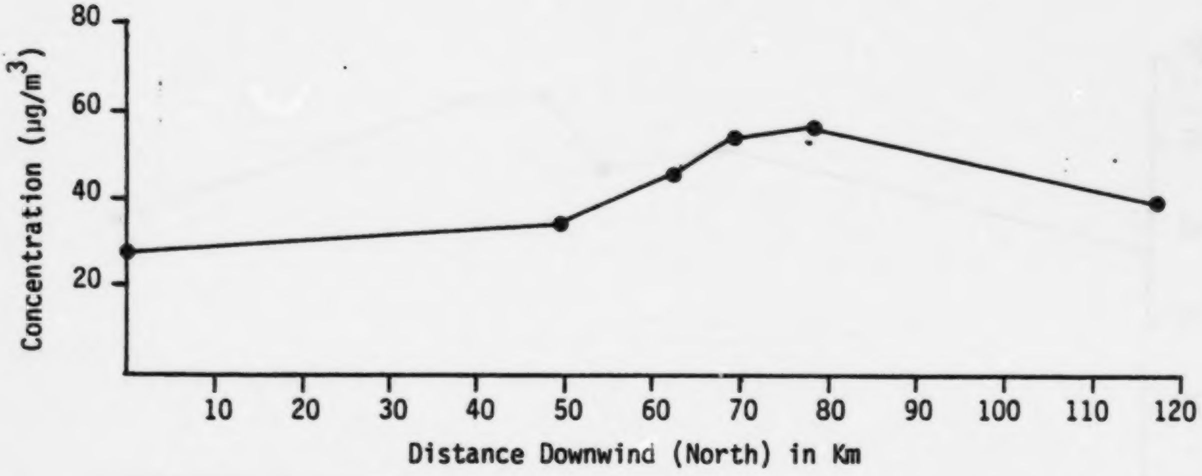

Figure 3.2b IP concentration cross-section, wind from south.

Figure 3.2 Regional upwind/downwind analyses for IP.

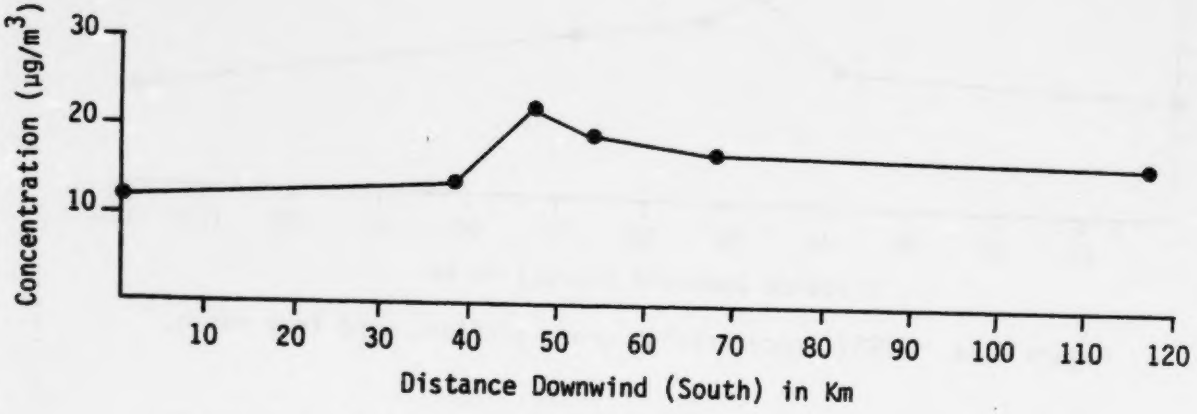

Figure 3.3a FINE concentration cross-section, wind from north.

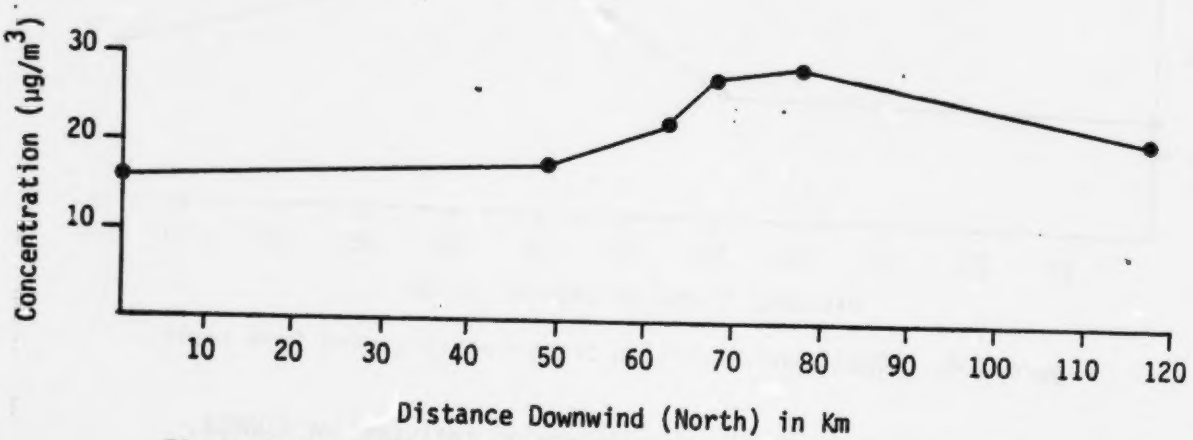

Figure 3.3b FINE concentration cross-section, wind from south.

Figure 3.3 Regional upwind/downwind analyses for FINE.

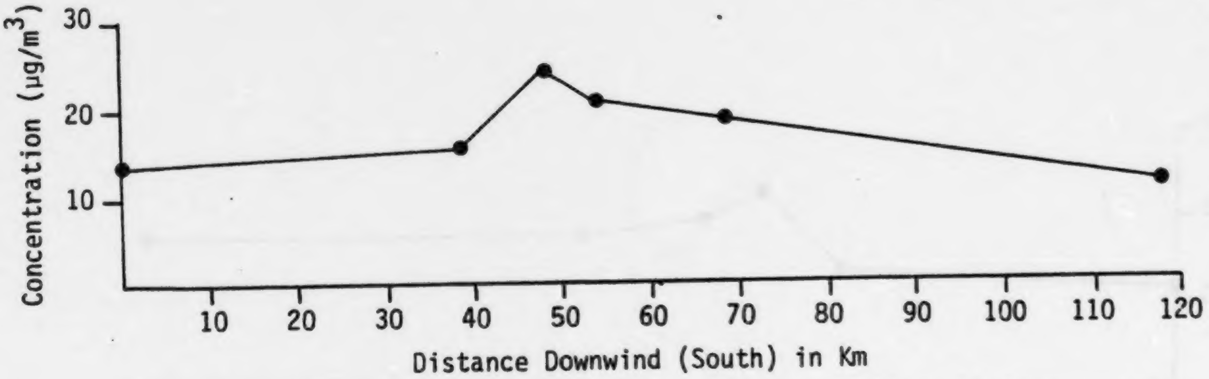

Figure 3.4a COARSE concentration cross-section, wind from north.

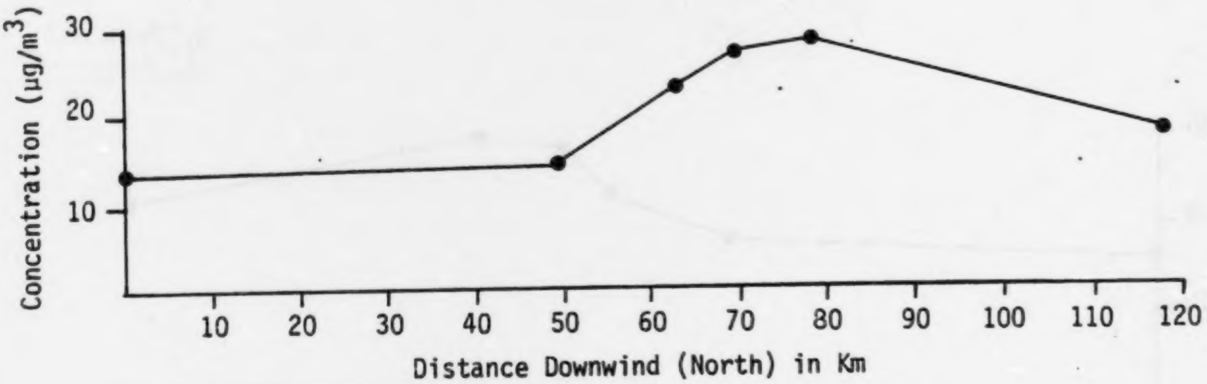

Figure 3.4b COARSE concentration cross-section, wind from south.

Figure 3.4 Regional upwind/downwind analyses for COARSE.

TABLE 3.1 SUMMARY OF UPWIND/DOWNWIND ANALYSES FOR TSP, IP, FINE, AND COARSE.

	AEROSOL CONCENTRATIONS ($\mu g/m^3$)							
	TSP		IP		FINE		COARSE	
Wind Direction	N	S	N	S	N	S	N	S
Average Concentration								
122	44.9	59.0	25.7	39.8	12.1	21.9	13.6	17.9
103	71.5	89.2	46.7	54.8	22.8	28.0	23.9	26.8
124	45.4	56.3	27.2	29.3	16.1	16.6	11.1	12.7
Downwind-Upwind*								
Absolute Change	0.5	2.7	1.5	10.5	4.0	5.3	-2.5	5.2
Percent Change	1%	5%	6%	36%	33%	32%	-18%	41%
Urban-Upwind**								
Absolute Change	26.6	32.9	21.0	25.5	10.7	11.4	10.3	14.1
Percent Change	59%	58%	82%	87%	88%	69%	76%	111%

*"Downwind-Upwind" is difference between downwind and upwind averages.

**"Urban-Upwind" is the difference between the site 103 reading and upwind rural site concentration.

1 to 3 μg/m^3. There exists, however, what might be called an "indirect" transport effect that cannot be measured by the regional upwind/downwind analysis. The indirect effect, which should be particularly important for secondary aerosols, involves sloshing of air masses in the AQCR during periods of light or variable winds. Making an allowance for indirect transport, we conjecture that the total impact of the St. Louis urban area on annual mean concentrations at the rural sites is on the order of 2 - 5 μg/m^3 of (mostly fine) particulate matter.

Figures 3.5 through 3.7 illustrate concentration profiles for sulfur, lead, and silicon. The first two elements, which are associated with FINE, do show a positive transport effect downwind of the city for both northerly and southerly flows. Silicon, which is associated with crustal material and COARSE, does not show a consistent transport effect. These results reinforce our above conclusion that the urban plume basically contains fine particles. The enrichment of the FINE/COARSE ratio in the urban plume compared to the center-city (where FINE and COARSE are about equal) likely results from two factors: (1) coarse particles settle during transport and (2) some fine particles (secondary aerosols) are produced during transport.

The urban versus rural concentrations of sulfur (or sulfate) deserve special comment. The annual mean concentration of ammonium sulfate (4.125 x sulfur) range only from 9.9 μg/m^3 at the lowest RAMS site (124) to 14.4 μg/m^3 at the highest RAMS site (103). That the rural concentrations are 70% of the urban concentrations implies that the sulfate phenomenon is air basin in scale if not synoptic in scale. This large spatial scale is rather typical for sulfates (EPA, 1975; Frank and Posseil, 1976) and occurs because sulfates are mostly secondary aerosols slowly produced from SO_x emissions (most of which arise from nonurban, tall-stack sources, i.e. power plants). The upwind/downwind analysis (averaged for northerly and southerly flows) yields the following averages for ammonium sulfate : upwind = 8.9 μg/m^3, center-city (103) = 12.3 μg/m^3, and downwind = 11.0 μg/m^3. This illustrates transport of sulfate from the urban center to downwind sites. More importantly, the upwind concentration being 70% of the center-city concentration

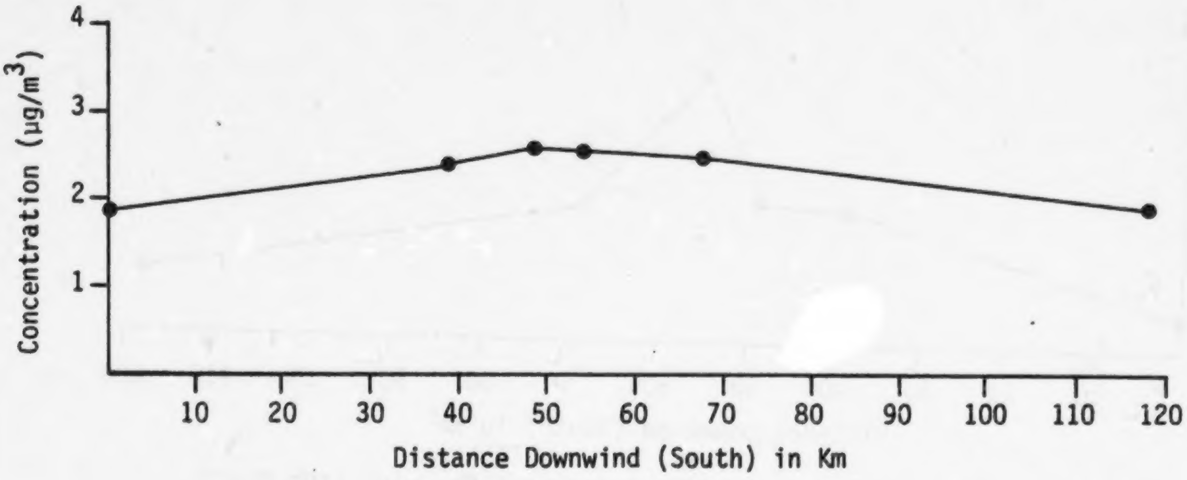

Figure 3.5a Sulfur concentration cross-section, wind from north.

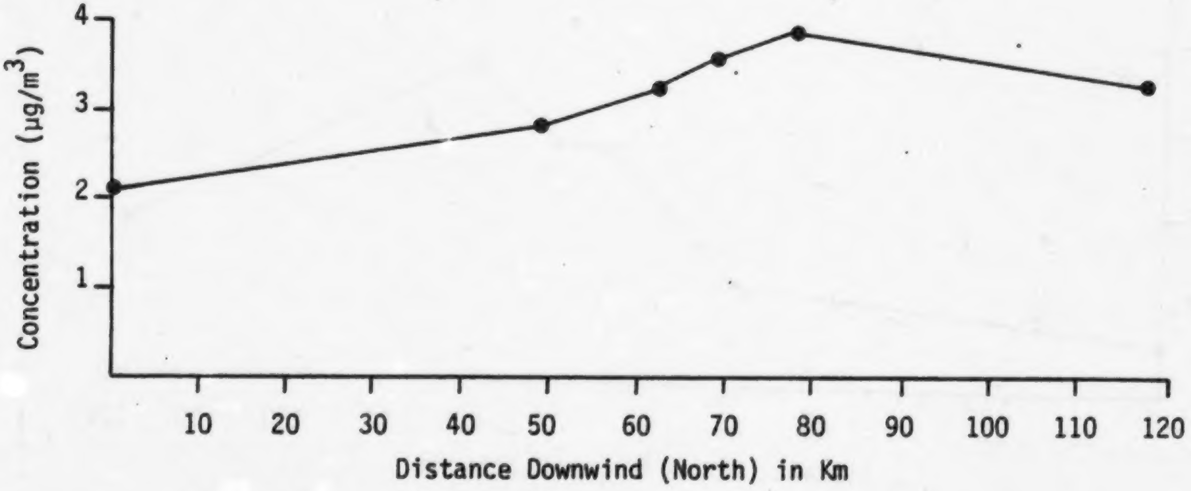

Figure 3.5b Sulfur concentration cross-section, wind from south.

Figure 3.5 Regional upwind/downwind analyses for sulfur.

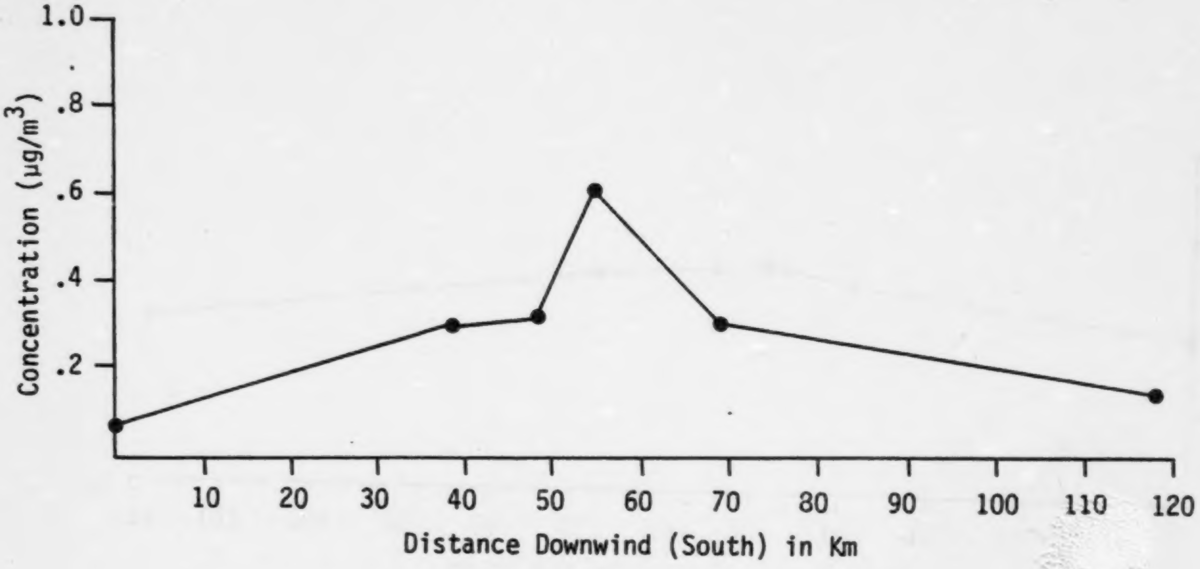

Figure 3.6a Lead concentration cross-section, wind from north.

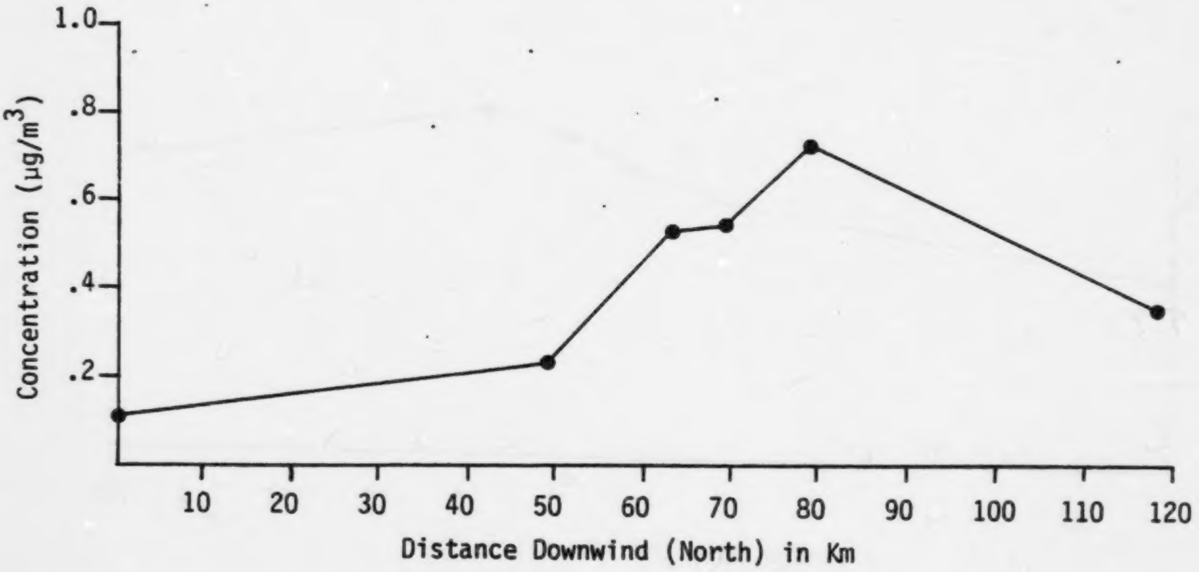

Figure 3.6b Lead concentration cross-section, wind from south.

Figure 3.6 Regional upwind/downwind analyses for lead.

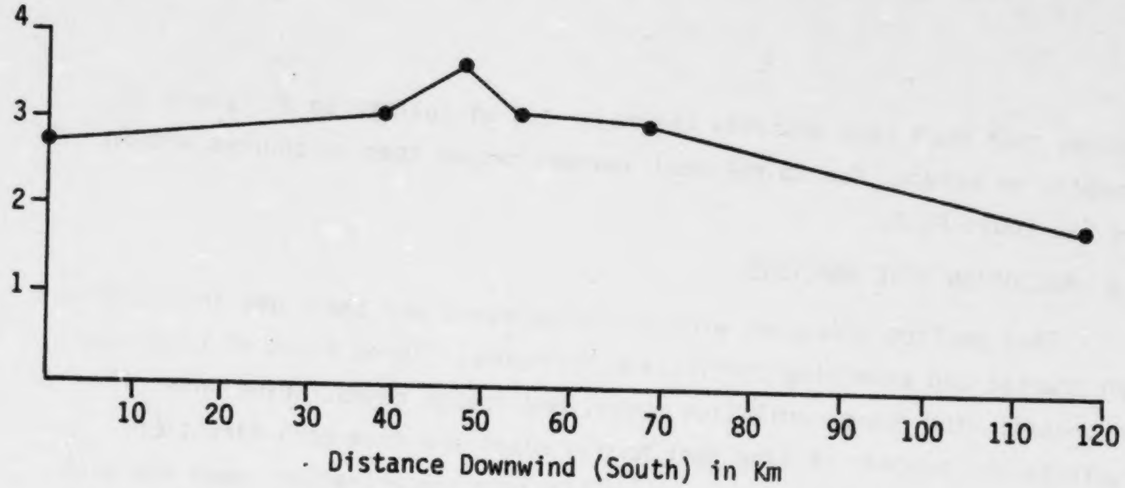

Figure 3.7a Silicon concentration cross-section, wind from north.

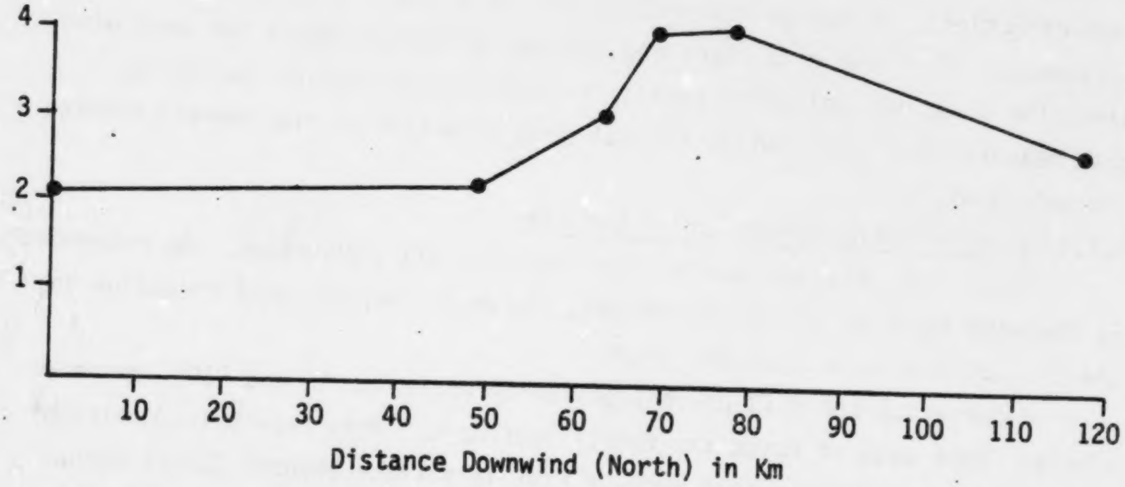

Figure 3.7b Silicon concentration cross-section, wind from south.

Figure 3.7 Regional upwind/downwind analyses for silicon.

implies that much (and possibly the majority) of sulfate in St. Louis is synoptic in nature, due to regional sources rather than to sources within the St. Louis AQCR.

3.2 POLLUTION ROSE ANALYSIS

This section discusses wind/pollution roses and their use in identifying sources and examining particulate transport. Three types of roses are presented: wind roses, pollution roses, and dosage roses. Wind roses illustrate the percent of time that hourly winds are from each direction. Pollution roses illustrate average particulate concentrations when the wind is from given directions.* Theoretically, the pollution roses for an atmospheric constituent emitted from a single source should all point approximately toward that source. In contrast, pollution roses for a constituent emitted or produced throughout a broad area should be isotropic (invarient with direction). A dosage rose represents the product of wind direction frequencies (i.e. the wind rose) and average pollution levels for each wind direction (i.e. the pollution rose); for each wind direction the dosage rose measures the contribution of that wind direction to the overall average concentration.

3.2.1 Wind/Pollution Roses for IP and FINE

Figure 3.8 presents hourly wind roses for the RAMS sites. As evidenced by the wind roses at all ten locations, the most frequent wind direction in the St. Louis area is from the south.

Figures 3.9 and 3.10 present pollution roses for IP and FINE, respectively. Both sets of roses are fairly isotropic. This result is consistent with earlier conclusions that much of FINE is sulfate aerosol (which occurs on a large spatial scale), and that the two dominant sources of IP are sulfates and ubiquitous fugitive dust sources. Both sets of roses do, however, appear to show some directional biases toward the industrial areas in East St. Louis and along the Mississippi River. Furthermore, the pollution roses for FINE seem generally to have a slight but detectable south to east bias (the reason for this will be seen in the next subsection).

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*In this chapter, we use the six-hour dichotomous data at sites 103 and 105 and the twelve-hour dichotomous data at other sites to construct pollution roses.

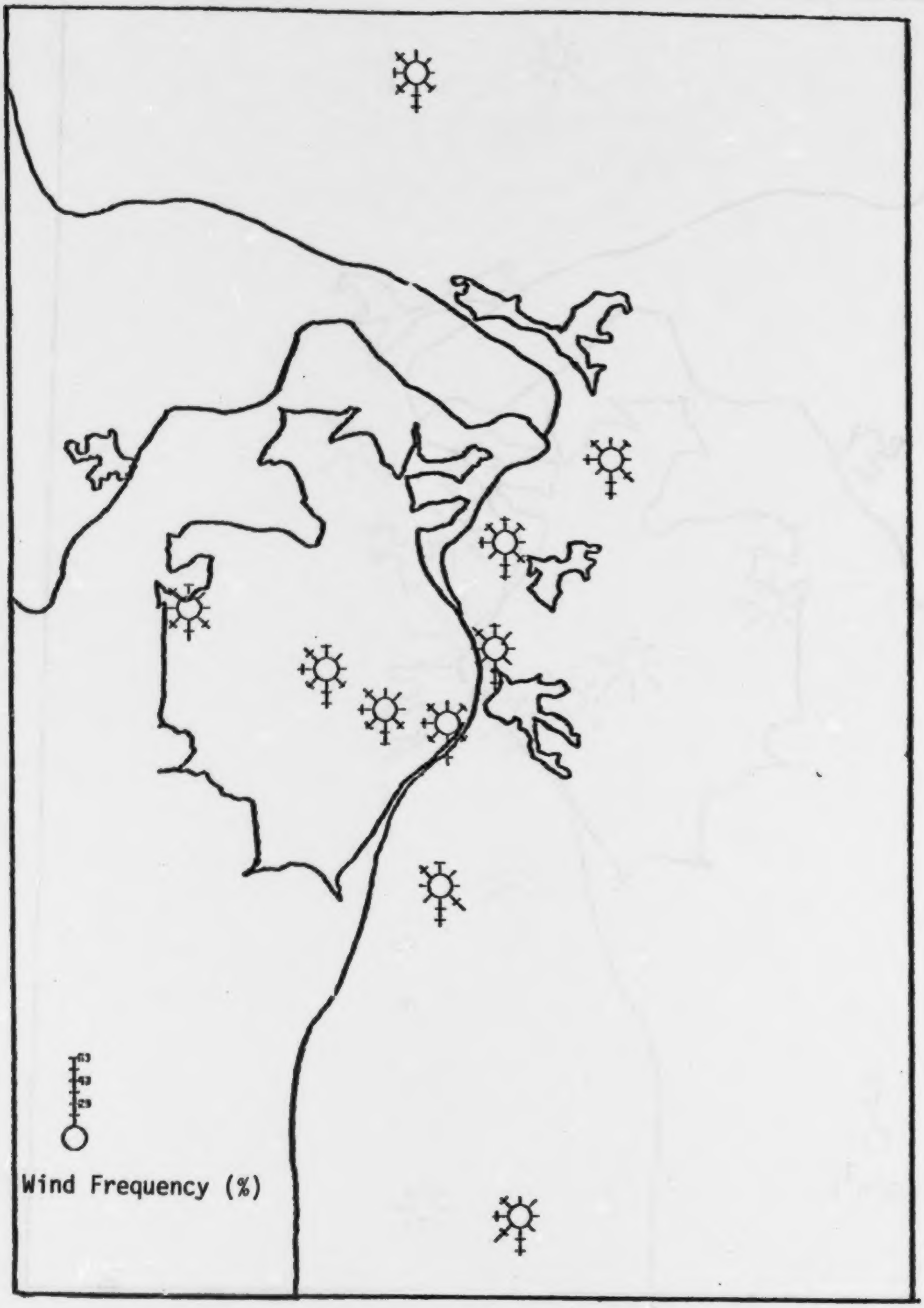
Wind Frequency (%)

Figure 3.8 Wind roses for the ten RAMS particulate sites.

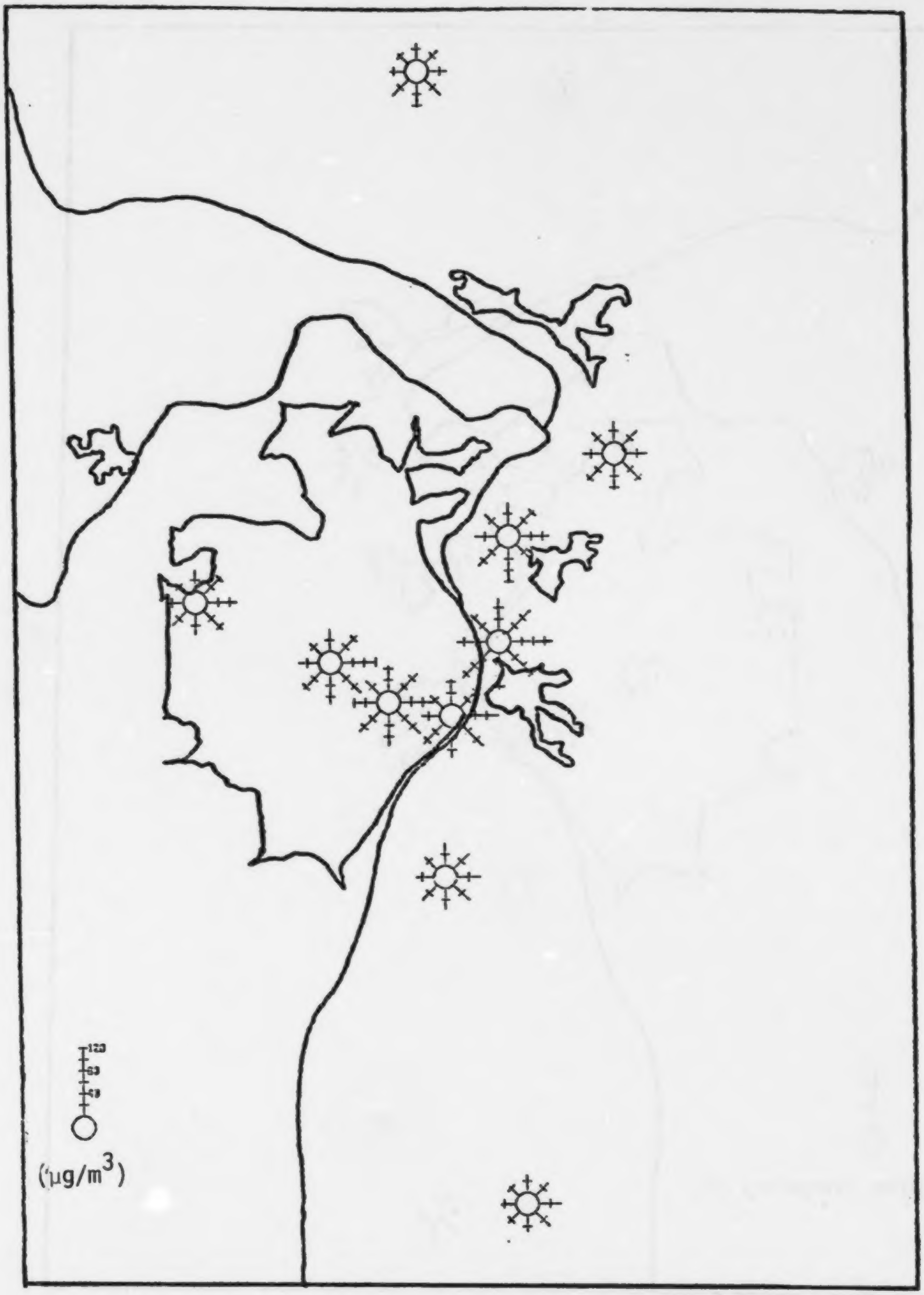
120
80
40
(µg/m³)

Figure 3.9 Pollution roses for IP at the ten RAMS particulate sites.

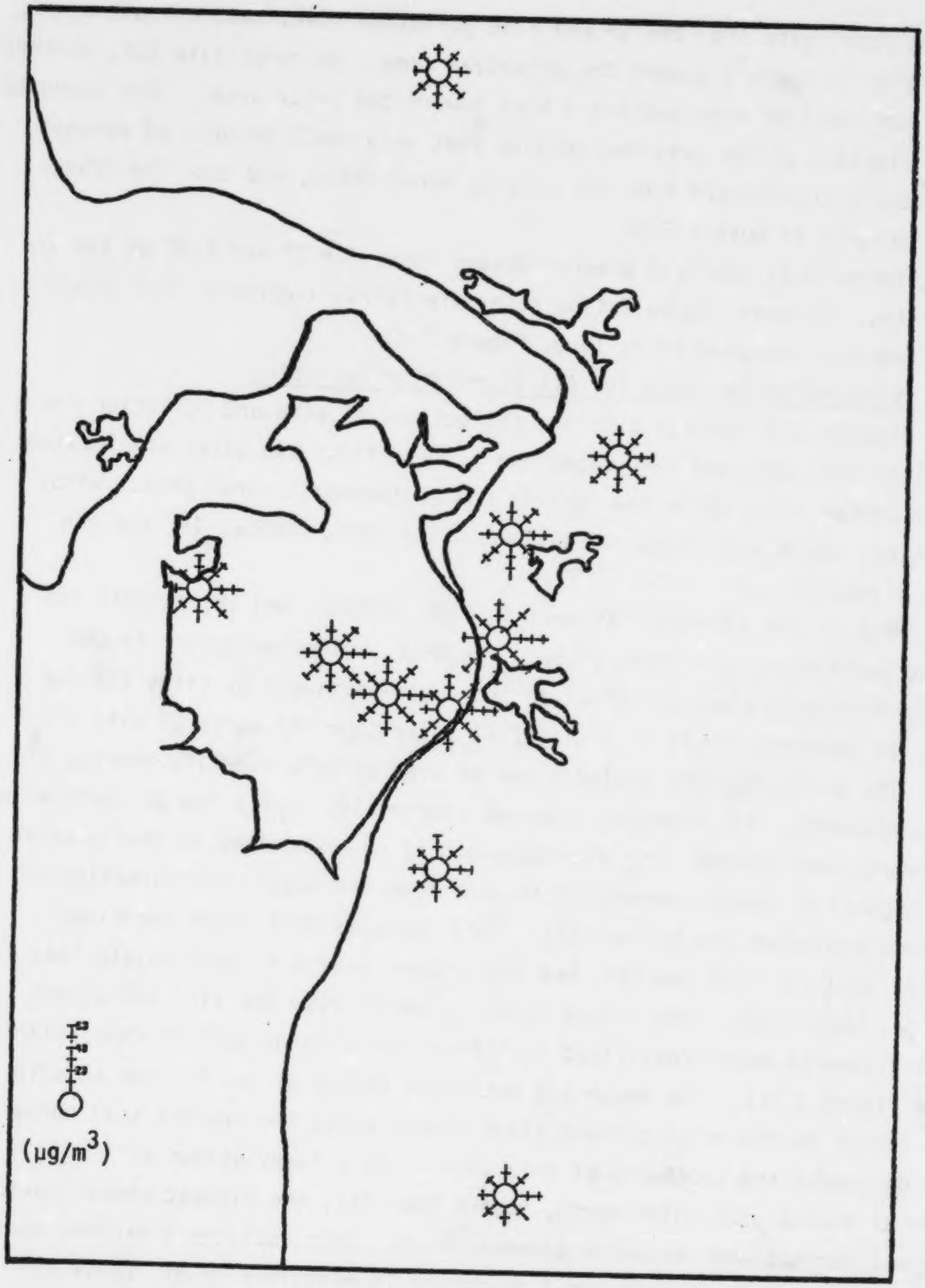

Figure 3.10 Pollution roses for FINE at the ten RAMS particulate sites.

At rural site 122, the IP and FINE pollution roses both exhibit a slight bias ($\sim 3\ \mu g/m^3$) toward the urbanized area. At rural site 124, neither the IP nor the FINE rose exhibit a bias toward the urban area. This supports the conclusions of the previous section that only small amounts of aerosol are directly transported from the city to rural sites, and that the transported aerosol is mostly FINE.

Figures 3.11 and 3.12 present dosage roses for IP and FINE at the ten RAMS sites. Because the pollution roses are fairly isotropic, the dosage roses resemble the wind roses (i.e. Figure 3.8).

3.2.2 Wind/Pollution Roses for All Dichotomous Variables

Figures 3.13 through 3.15 are collections of wind and pollution roses for sites 103, 105, and 124 respectively. The first two sites are located in the center city, while the last is the southernmost rural site. Wind, pollution, and dosage roses are presented for FINE, COARSE, IP, and the eight elements.

Most of the elements (as well as FINE, COARSE, and IP) exhibit isotropic patterns at all three sites. The most obvious exception is the highly wind-biased pattern of Ti to the south-southwest at sites 103 and 105. In contrast, there is a slight Ti spike from the north at site 124.

The pollution rose analysis can be used to help identify sources of trace elements. For example, although station 103 lies a few km south of a secondary lead smelter, its Pb rose exhibits no pronounced northerly spike. The highest Pb levels correspond to wind from the south, the direction of maximum vehicular traffic density. This suggests that motor vehicles, rather than the lead smelter, are the primary source of particulate lead in the St. Louis area. The strong spike of the Ti rose for site 103 points toward several major coal-fired facilities and a large paint pigment plant (see Figure 3.16). The south and southwest spikes of the Ti rose at site 105 are caused by the paint pigment plant alone, since the nearest coal burners lie northwest and southeast of this site. The primary spikes of the Ti rose at station 124 point north, toward the city, the highest concentration of coal burners and the paint pigment plant. This analysis indicated that the paint plant is the principal source of Ti emissions in St. Louis.

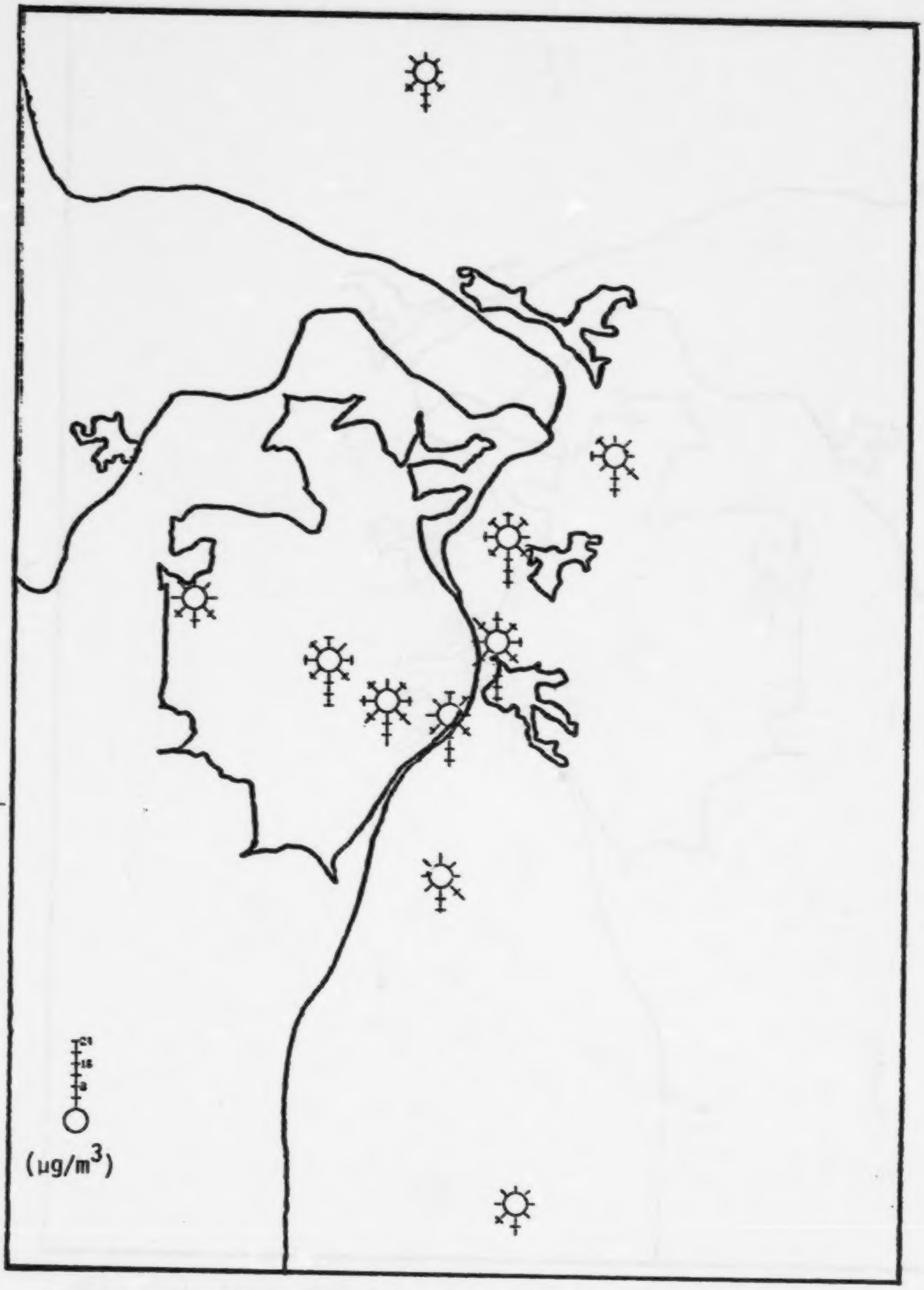

Figure 3.11 Dosage roses for IP at the ten RAMS particulate sites.

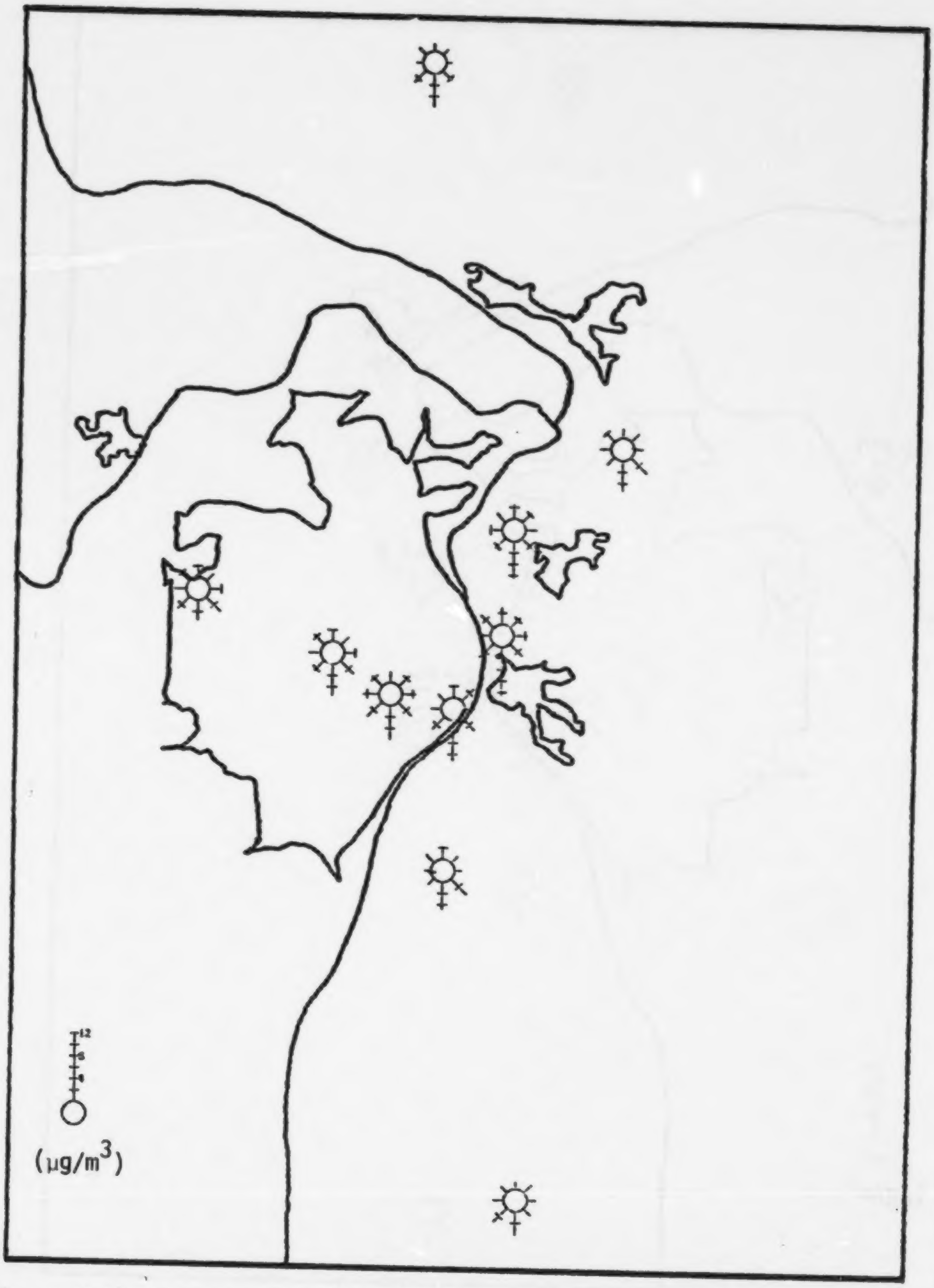

Figure 3.12 Dosage roses for FINE at the ten RAMS particulate sites.

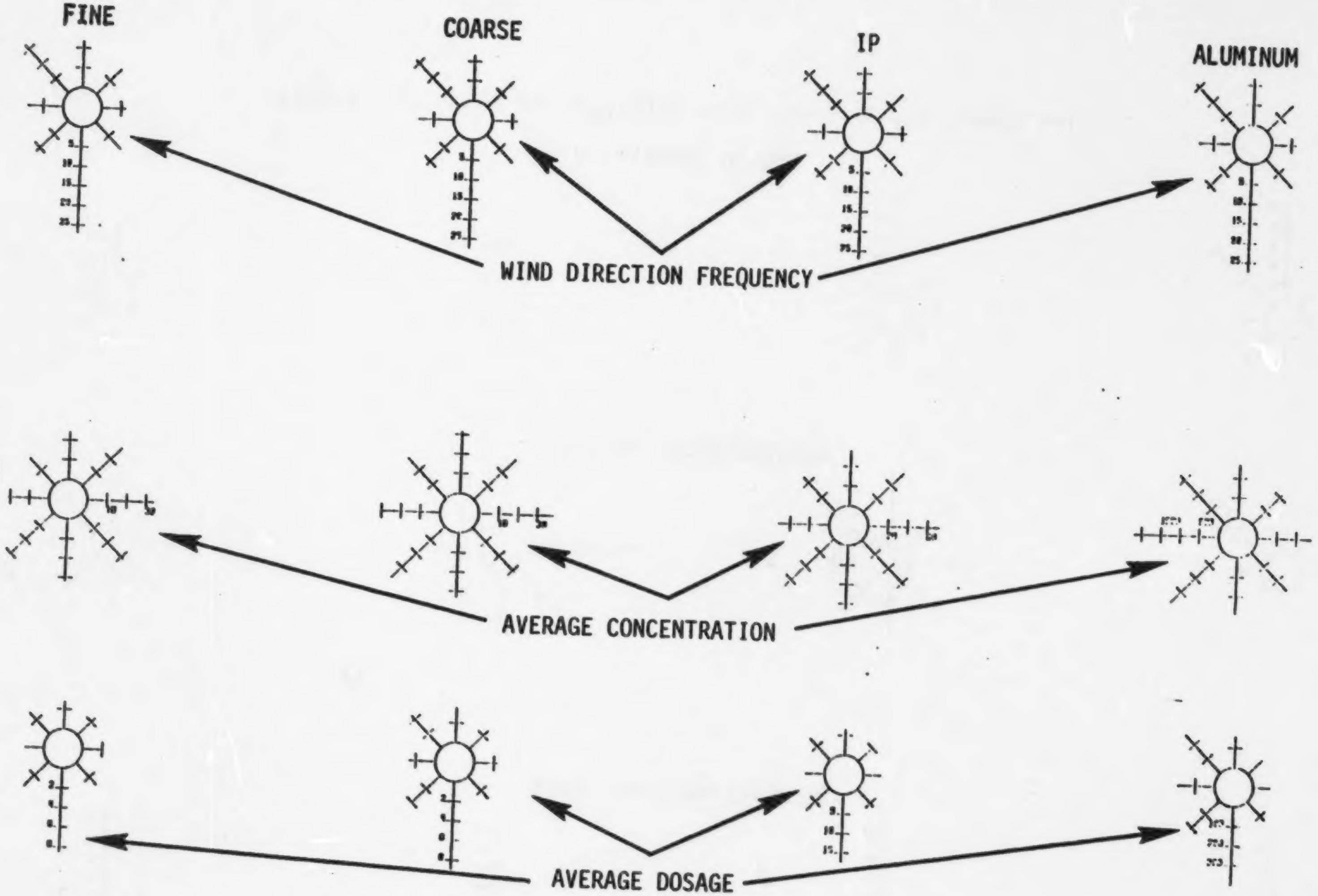

Figure 3.13 Wind and pollution roses for site 103.

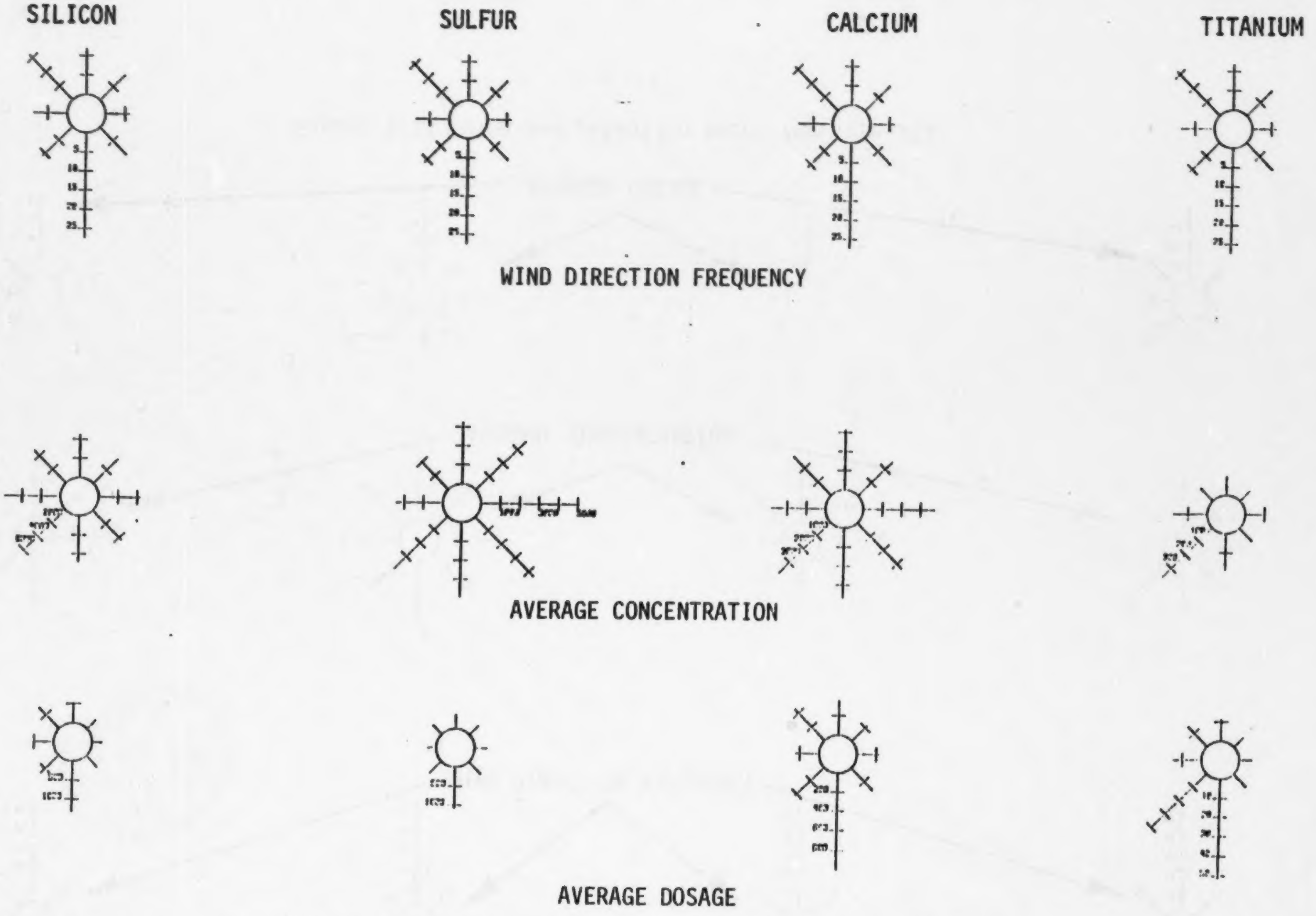

Figure 3.13 Wind and pollution roses for site 103 (continued).

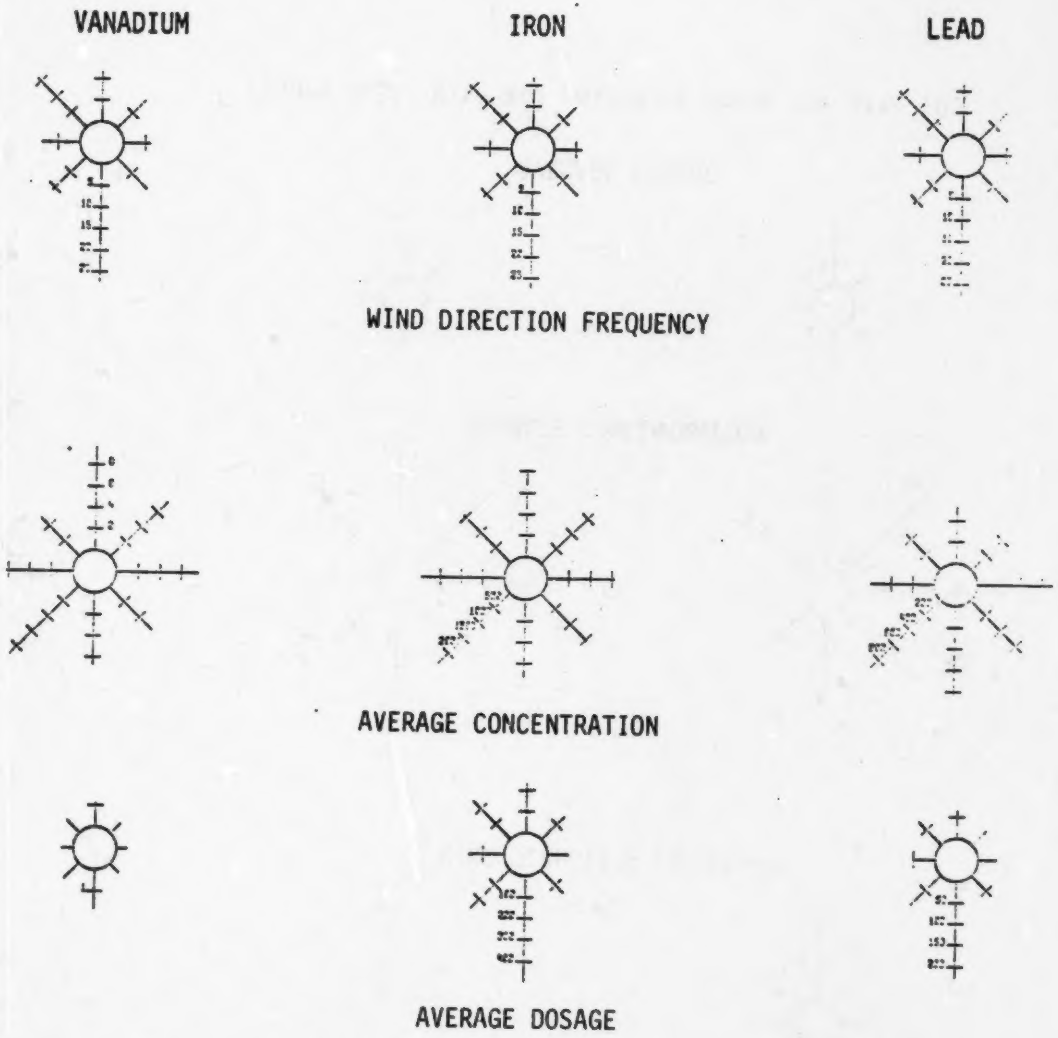

Figure 3.13 Wind and pollution roses for site 103 (continued).

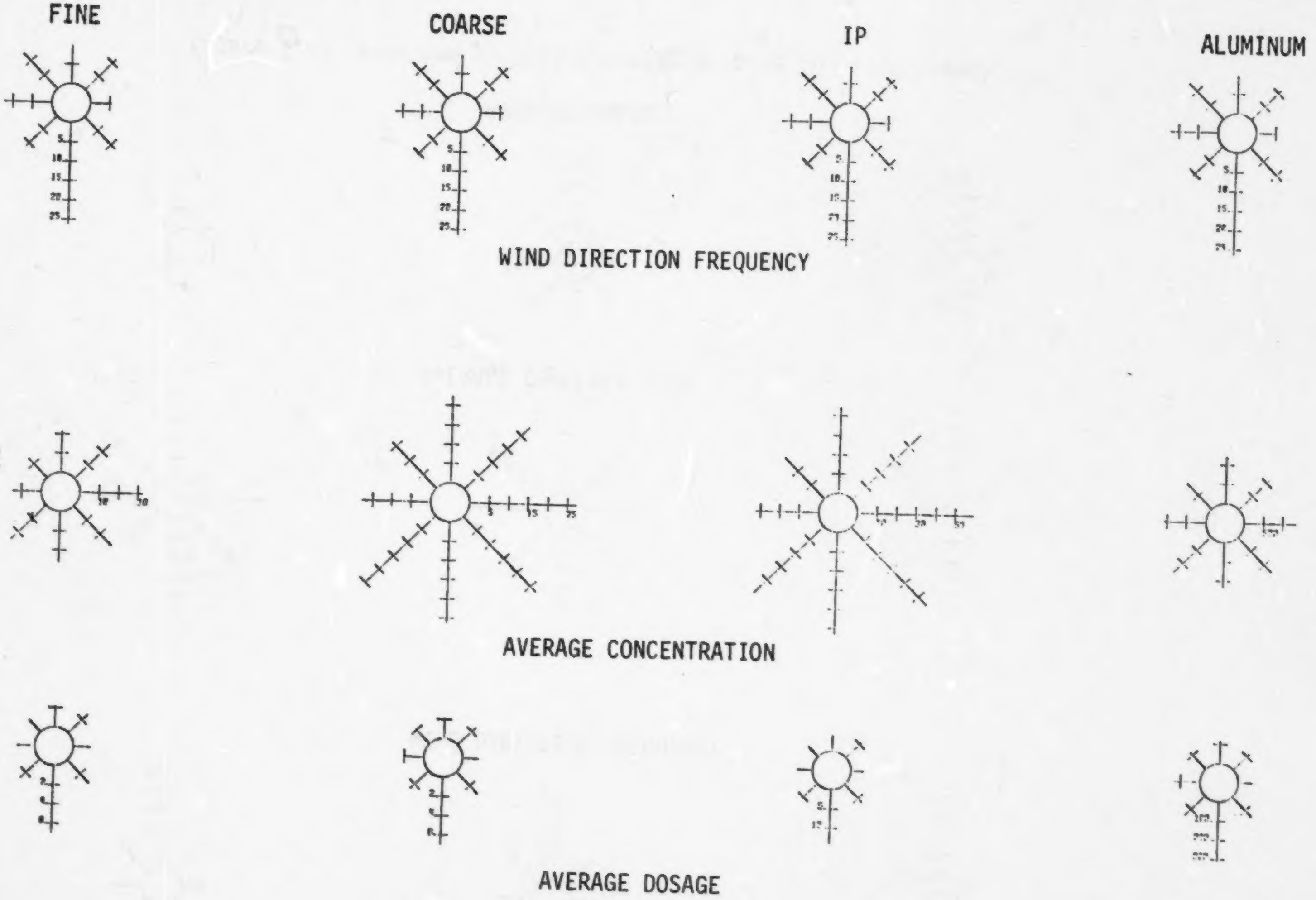

Figure 3.14 Wind and pollution roses for site 105.

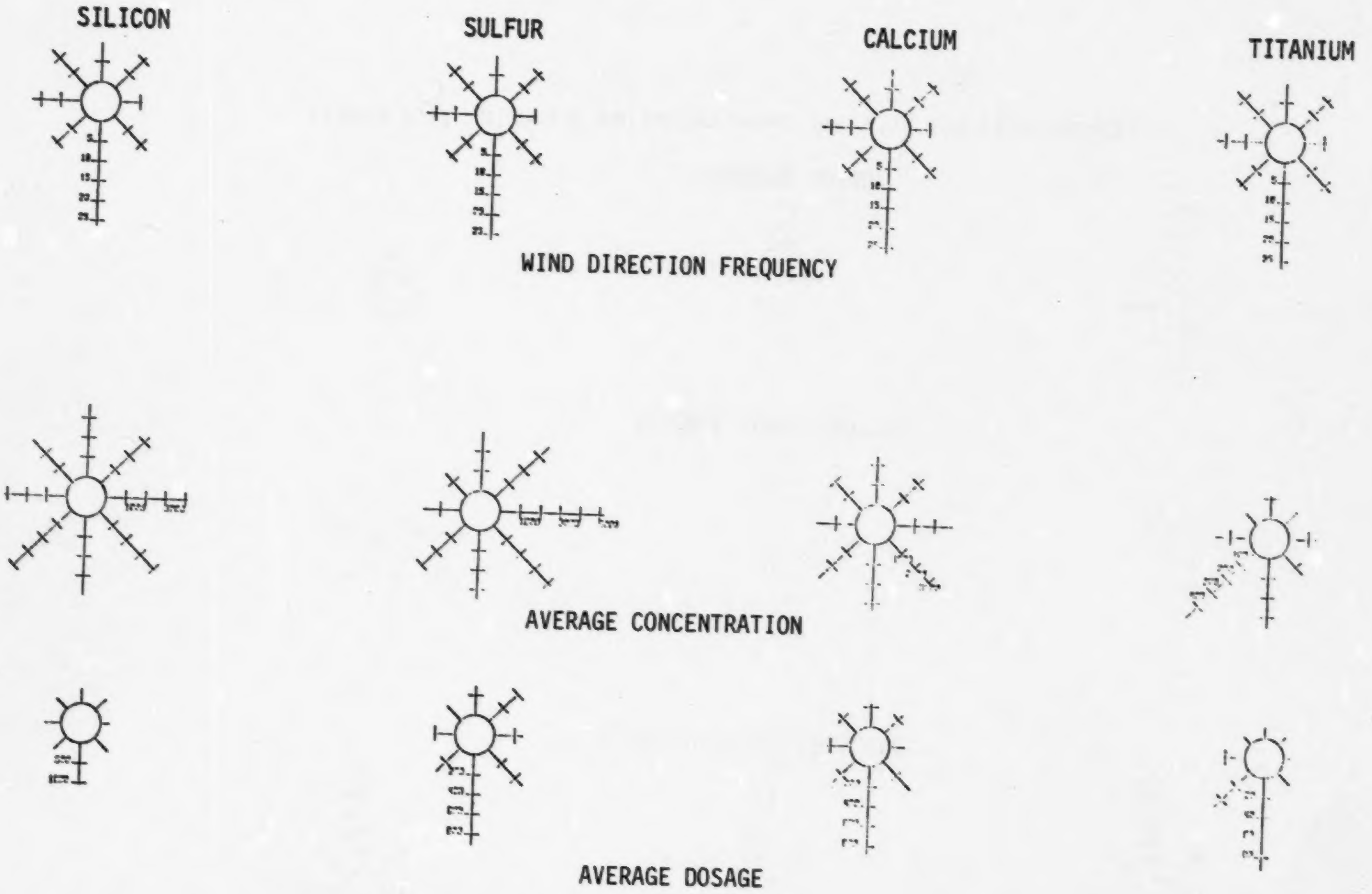

Figure 3.14 Wind and pollution roses for site 105 (continued).

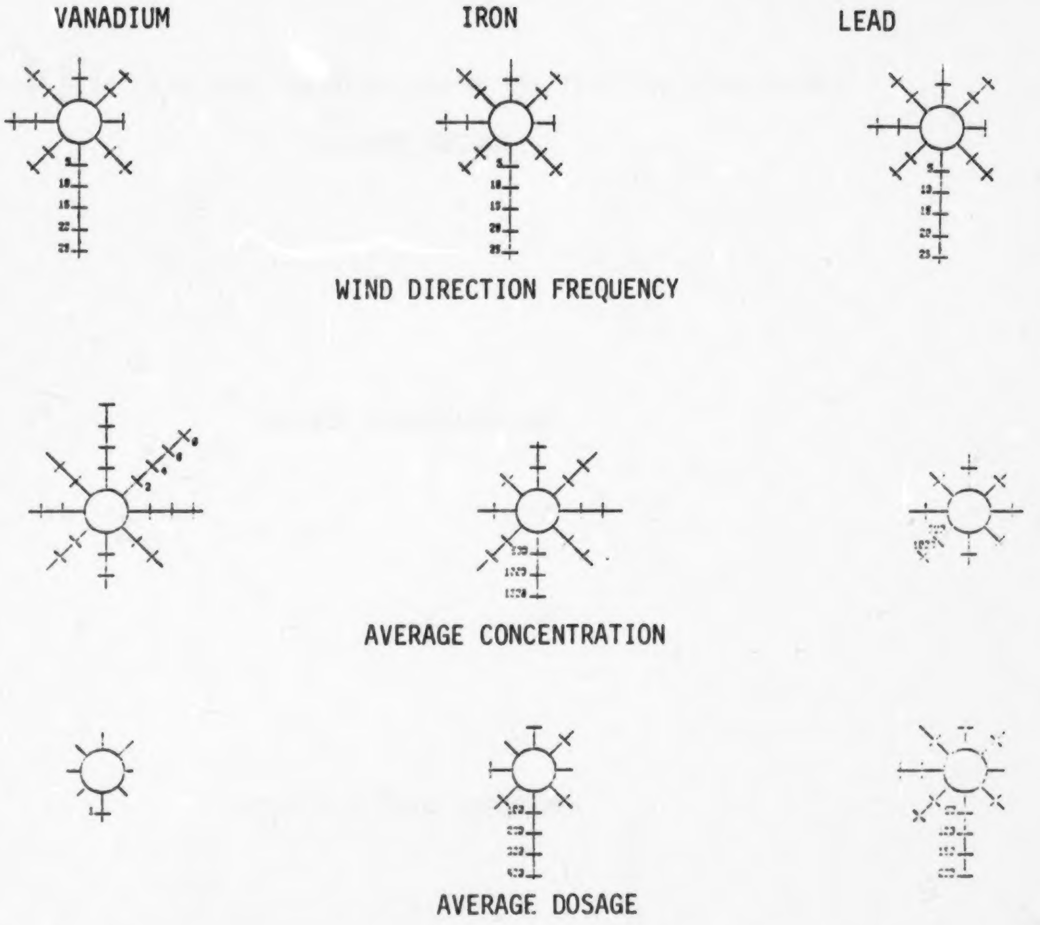

Figure 3.14 Wind and pollution roses for site 105 (continued).

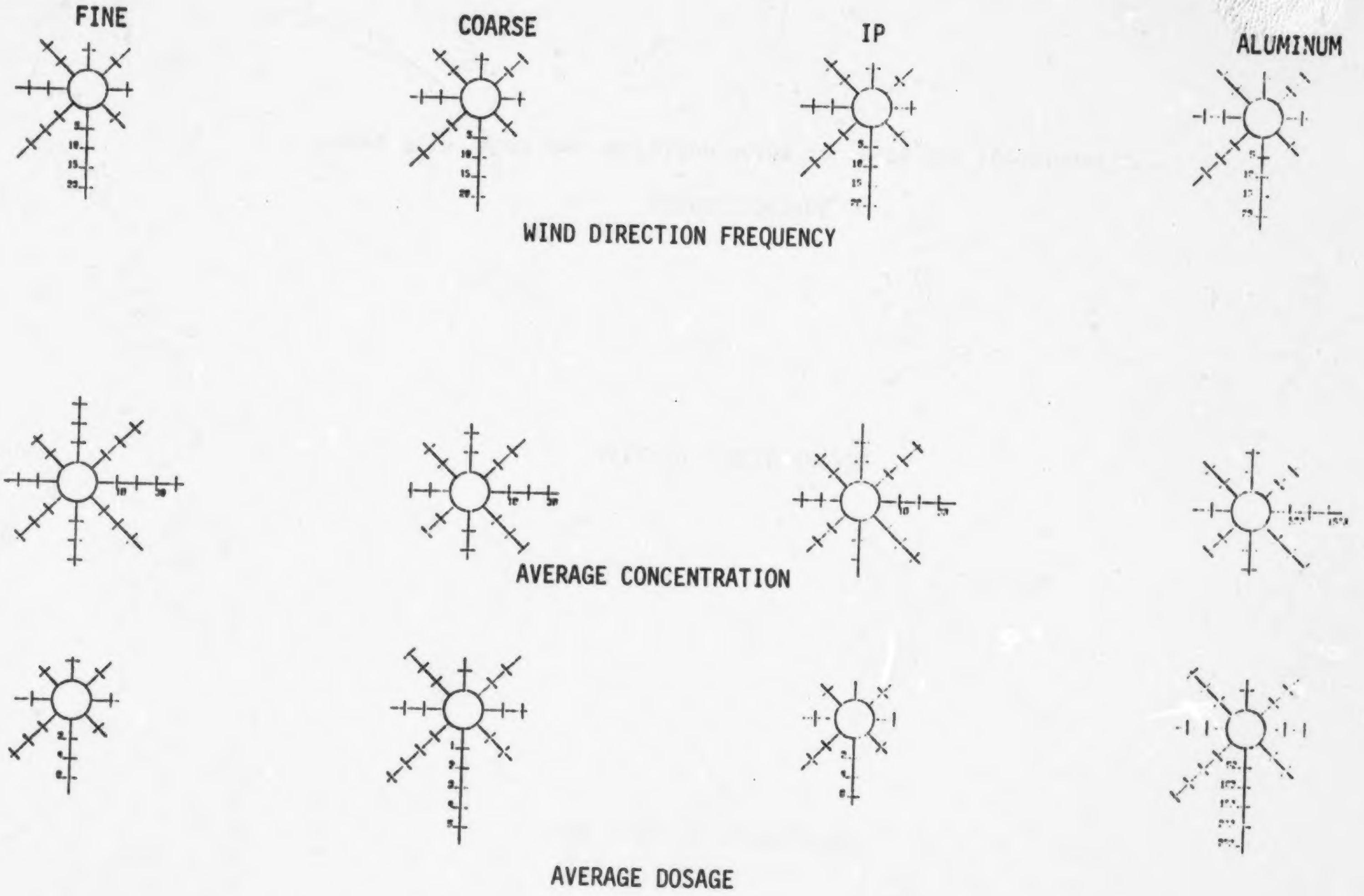

Figure 3.15 Wind and pollution roses for site 124.

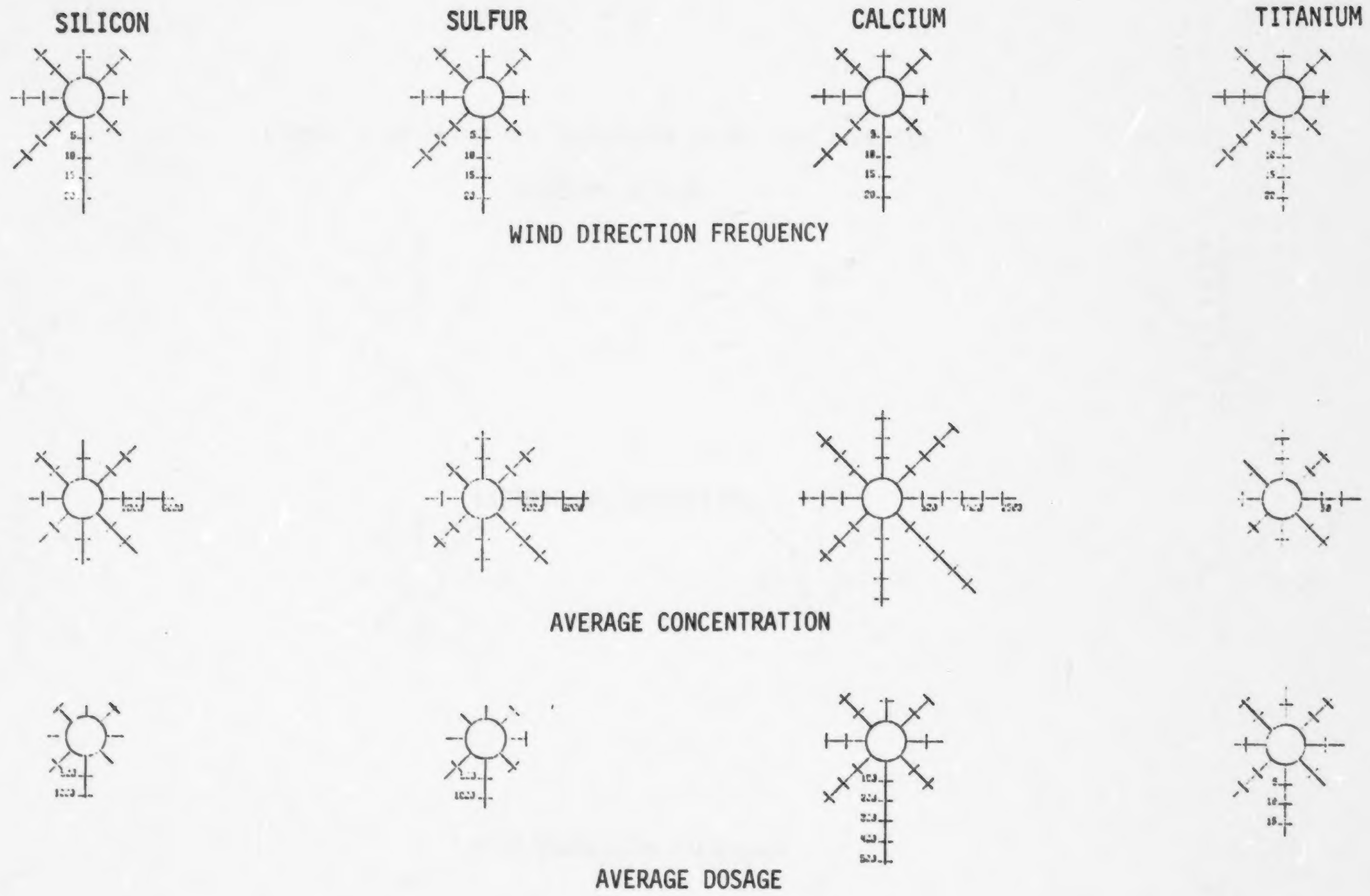

Figure 3.15 Wind and pollution roses for site 124 (continued).

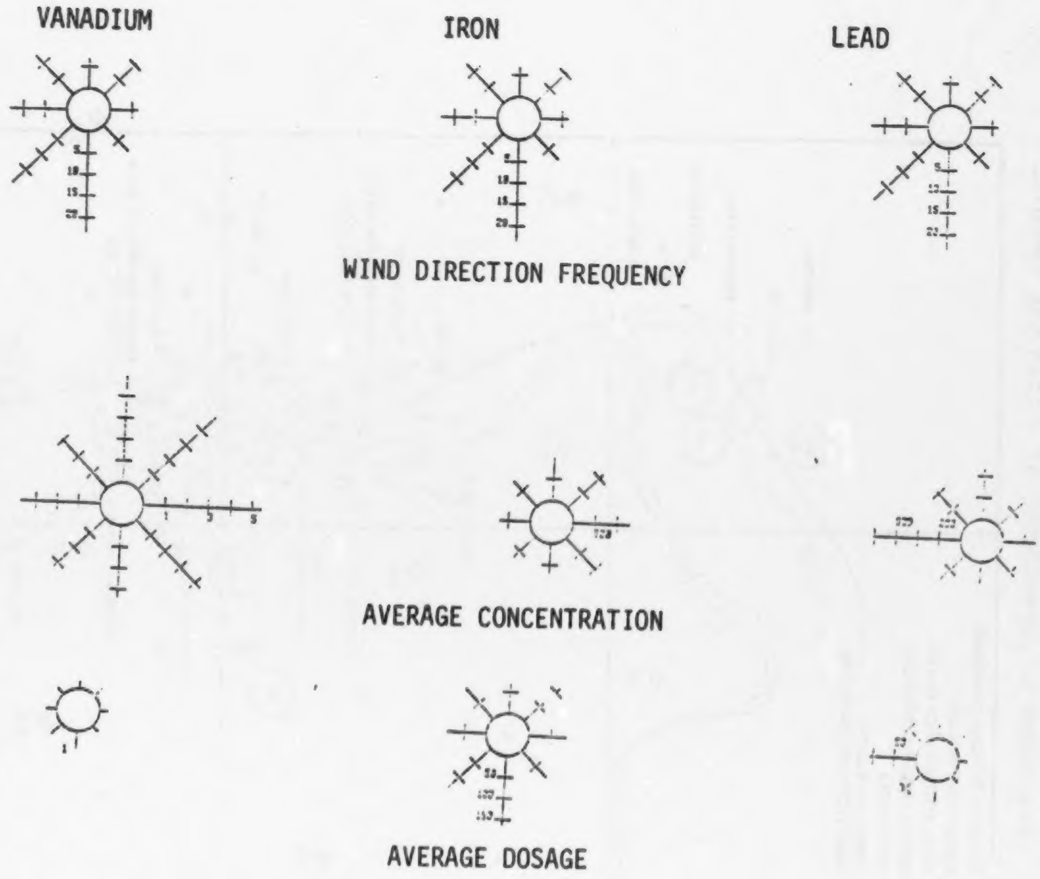

Figure 3.15 Wind and pollution roses for site 124 (continued).

Note: for selected sources, emission rates in tons/year
are shown in parentheses or within the symbols.

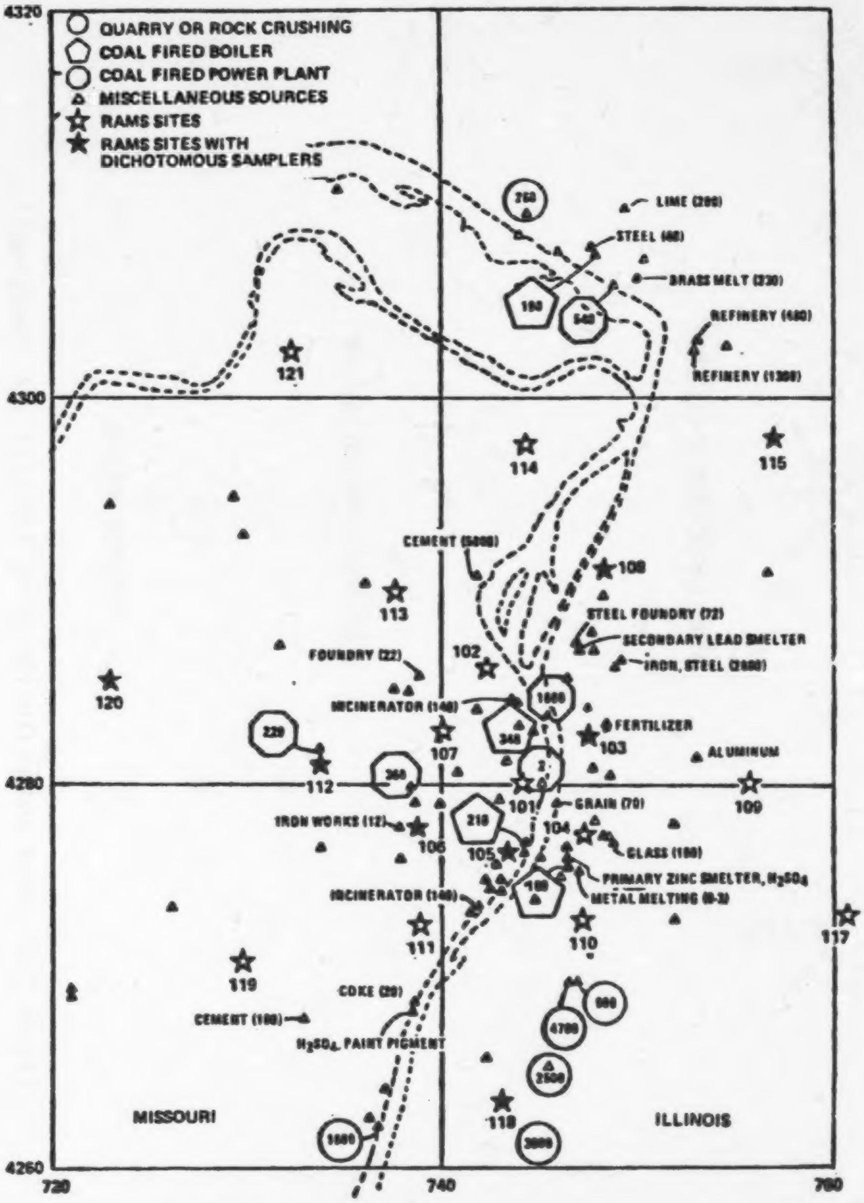
4320
QUARRY OR ROCK CRUSHING
COAL FIRED BOILER
COAL FIRED POWER PLANT
MISCELLANEOUS SOURCES
RAMS SITES
RAMS SITES WITH DICHOTOMOUS SAMPLERS
250
LIME (200)
STEEL (40)
BRASS MELT (230)
190
540
REFINERY (480)
REFINERY (1300)
121
4300
114
115
CEMENT (5000)
108
113
STEEL FOUNDRY (72)
SECONDARY LEAD SMELTER
FOUNDRY (22)
102
IRON, STEEL (2000)
1600
INCINERATOR (140)
FERTILIZER
120
220
340
103
107
ALUMINUM
2
4280
112
360
101
109
GRAIN (70)
IRON WORKS (12)
210
104
106
GLASS (100)
105
PRIMARY ZINC SMELTER, H_2SO_4
METAL MELTING (0-3)
INCINERATOR (140)
180
117
111
110
119
900
COKE (20)
CEMENT (100)
4700
H_2SO_4, PAINT PIGMENT
2500
MISSOURI
118
ILLINOIS
1600
3000
4260
720
740
760

Figure 3.16 Emission sources in the vicinity of St. Louis
(Dzubay, 1979).

At all three sites, FINE and sulfur exhibit a moderate easterly to southerly bias. Because sulfate constitutes approximately 50% of FINE, the agreement between the pollution roses for sulfur and FINE is not unexpected. The east to south bias may reflect large scale sulfate phenomena, i.e. the dense SO_x emissions in the Ohio Valley toward the east, or high pressure systems moving from the south which are known to be conducive to sulfate formation.

The isoptropic nature of the pollution roses for COARSE and the crustal elements (Si, Al, and Ca) supports the contention made in Chapter 2 that the crustal material stems mostly from ubiquitous area sources of fugitive dust rather than point sources of fly ash. The isoptropic property also reflects the fact that the RAMS sites tend to be located away from strong local influences (local fugitive dust or other local sources).

3.3 ESTIMATE OF BACKGROUND CONCENTRATIONS

Annual mean background particulate concentrations for the St. Louis area can be calculated by taking average concentrations for the rural sites (122 and 124) and subtracting both transported aerosol from the urban area and local man-made aerosol. The results of the previous two sections indicate that the influence of the urban area on the rural sites amounts to approximately 2 to 5 $\mu g/m^3$ of essentially fine aerosol. Record et al. (1976) concluded that the local source influence on TSP at rural locations is on the order of 2 to 4 $\mu g/m^3$. The local influence should be basically coarse particles, and the mass contribution should be somewhat less for IP (or COARSE) than it is for TSP (because IP and COARSE do not include particles greater than 20 μm in size).

From the above considerations, we make an initial estimate that background aerosol concentrations for the St. Louis area are approximately 45 $\mu g/m^3$ for TSP, 25 $\mu g/m^3$ for IP, 13 $\mu g/m^3$ for FINE, and 12 $\mu g/m^3$ for COARSE. As noted previously in this chapter, these background concentrations represent natural sources plus man-made sources exterior to the St. Louis AQCR.

Record et al. (1976) estimated a background TSP concentration of approximately 30 μg/m^3 for the Midwest.* The large difference between their estimate and ours is probably due to three factors: (1) Record et al. seem to have too low of an estimate for transported secondary aerosols (only 3 μg/m^3); (2) the local scale contribution that we assumed, 2 to 4 μg/m^3 based on Record et al., may be too low (i.e. local scale contributions at the rural RAMS sites may be elevated due to fugitive dust from agriculture, roads, construction, etc.); and (3) the transport contribution from the urban area may be higher than we were able to detect with rather crude wind-direction analyses. Until these issues are resolved by further analysis, the background aerosol concentration for St. Louis must be regarded as somewhat uncertain. The best guess, however, based on current information would appear to be as follows:

TSP . . .	35 μg/m^3
IP . . .	20 μg/m^3
FINE . . .	10 μg/m^3
COARSE . . .	10 μg/m^3

---

*Actually, Record et al. (1976) estimated a background contribution of 25 μg/m^3 in terms of annual geometric mean (AGM). By examining data at non-urban locations, we have found that a 25 μg/m^3 AGM typically corresponds to a 30 μg/m^3 arithmetic average.

4.0 TEMPORAL PATTERNS OF ST. LOUIS PARTICULATE DATA

In response to seasonal and diurnal patterns of meteorology as well as to seasonal, weekly, and diurnal patterns of human activities, ambient aerosol concentrations exhibit significant temporal variations. These temporal patterns can frequently provide insight into the nature and sources of particulate matter. This chapter discusses the temporal patterns of several meteorological variables and several components of particulate, as monitored at Lambert Field and the ten RAMS particulate sites.

4.1 SEASONAL PATTERNS

This section discusses the seasonal patterns of twelve particulate parameters and eight meteorological parameters. The particulate and weather patterns are then analyzed jointly to assess the effects of meteorology on ambient aerosol concentrations.

4.1.1 Particulate Parameters

Figure 4.1 illustrates the seasonal patterns of TSP, IP, COARSE, and FINE. Each pattern consists of monthly averages for the year 1976. The data are grouped according to four urban sites (103, 105, 108, and 112), three suburban sites (115, 118, and 120), and one rural site (122).[*] As noted in Chapter 2, urban concentrations are higher than suburban concentrations which in turn are higher than rural concentrations. Suburban levels are generally closer to rural levels than urban levels.

All four particulate parameters in Figure 4.1 tend to peak during the summer (June through September). The summer peak is strong for FINE, moderately strong for IP, moderate for COARSE, and very weak for TSP.

Figure 4.2 displays the seasonal patterns for the eight elements. The urban versus suburban versus rural differences are again obvious. As noted in Chapter 3, the spatial differences are especially pronounced for Pb, V, Ti, and Fe and least pronounced for S.

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[*] Sites 105 and 124 are excluded from the seasonal analysis because dichotomous measurements were not taken at these sites from September through December.

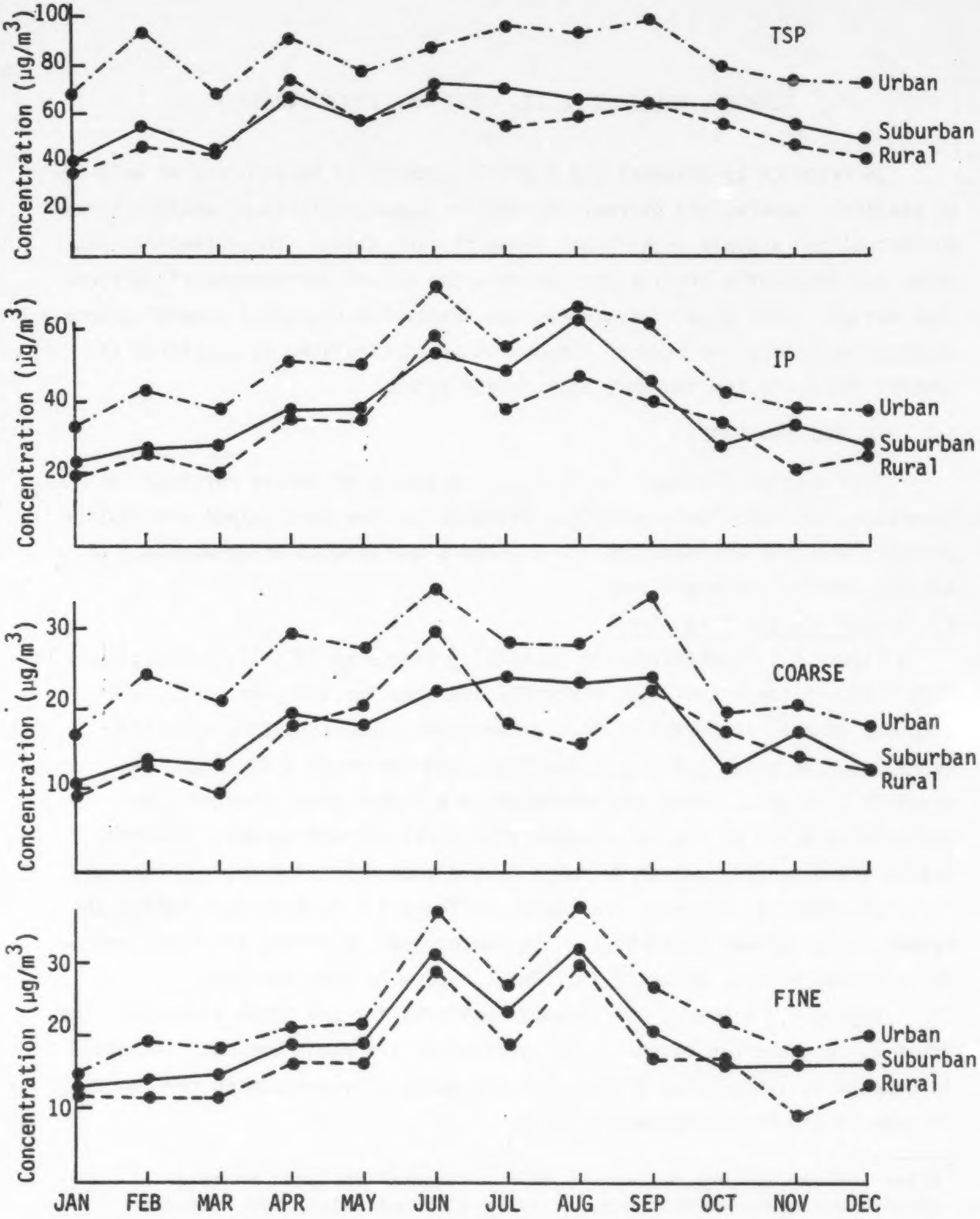

Figure 4.1 Seasonal patterns of TSP, IP, COARSE, and FINE.

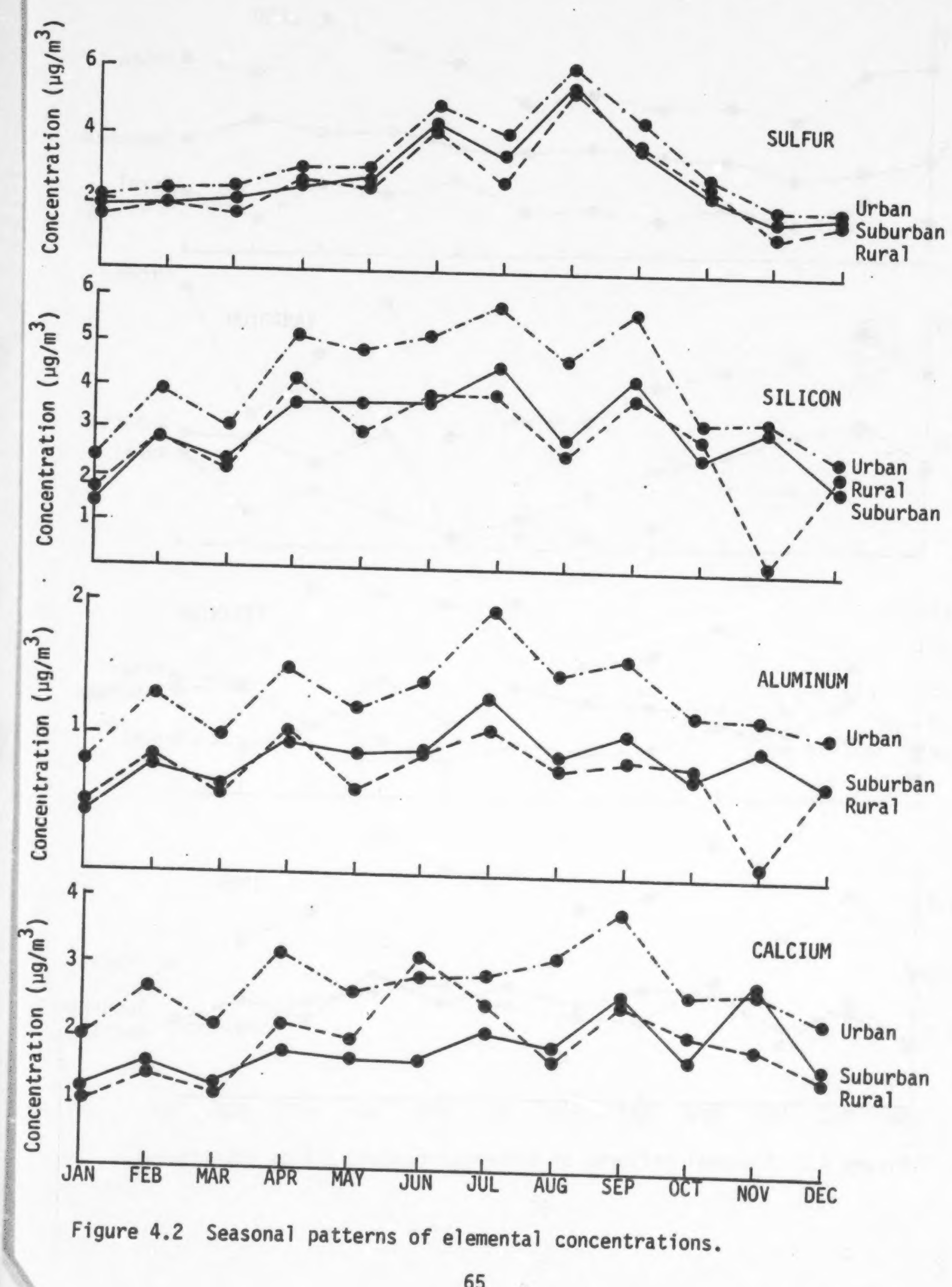

Figure 4.2 Seasonal patterns of elemental concentrations.

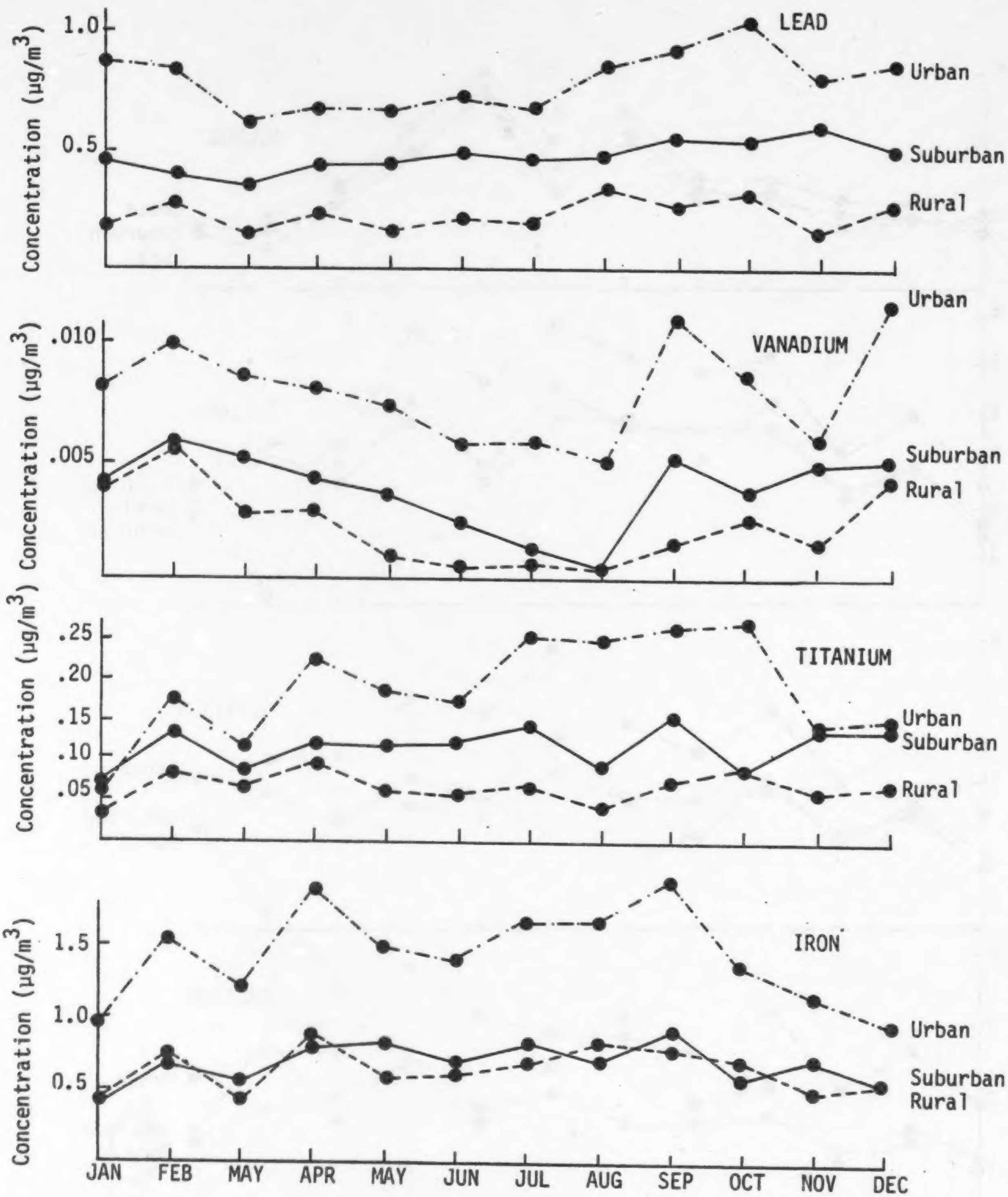

Figure 4.2 Seasonal patterns of elemental concentrations (continued).

Figure 4.2 indicates that sulfur has a strong summer peak, similar to the pattern of FINE. The parallel behavior of sulfur and FINE, a phenomenon noted throughout this report, is consistent with the results of the chemical element balance (Chapter 1) that showed sulfate contributes about one-half of FINE.

The crustal elements -- Si, Al, and Ca -- display moderate summer peaks, similar to COARSE. This is consistent with other findings in this report that crustal material constitutes the dominant part of COARSE. Titanium and iron show seasonal patterns fairly similar to those for Si, Al, and Ca, indicating that crustal material (in addition to the paint pigment plant and iron/steel mills) may be a major source of Ti and Fe as well.

The tendency for vanadium to peak in the winter reflects increased use of fuel oil for space heating during the winter. The element Pb, appears to lack a significant seasonal pattern. This evidently implies that gasoline consumption, as well as weather conditions governing the dispersion of lead, tend to be fairly constant during the year. An alternative explanation, that traffic and weather correlate inversely, such that periods of high vehicle use (e.g. summer) are accompanied by favorable (dispersive) meteorology, seems ruled out by the weather pattern studies of the next section.

4.1.2 Meteorological Parameters

Figure 4.3 illustrates the seasonal trend in daily average wind speed, showing a peak in the winter and a trough in the summer. Not surprisingly, Figure 4.4, the average value of number days persistent calm, shows an inverse pattern, peaking in the summer with a minimum during the winter.

Figure 4.5 shows that afternoon mixing height reaches a maximum in the summer months, falling to a minimum during the winter. Morning mixing height exhibits a less pronounced and inverse pattern, with a summer minimum. The daily spread between morning and afternoon mixing height, a measure of the amount of ground heating by the sun, is highest in summer, as one would anticipate.

Figure 4.6 shows the expected seasonal trends in daily minimum and maximum temperatures, with highs in the summer and lows in the winter. The

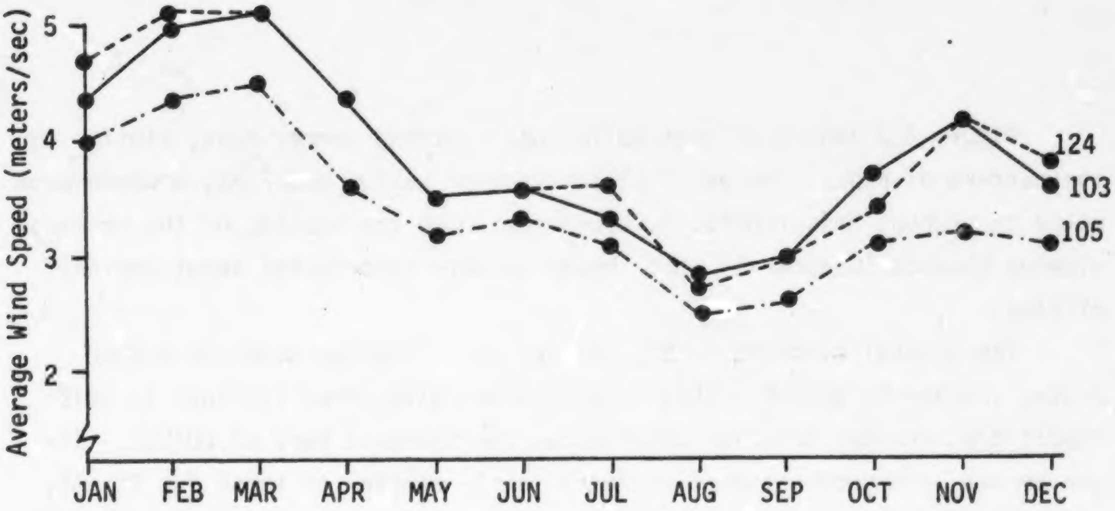

Figure 4.3 Monthly averages of daily (vector) average wind speeds.

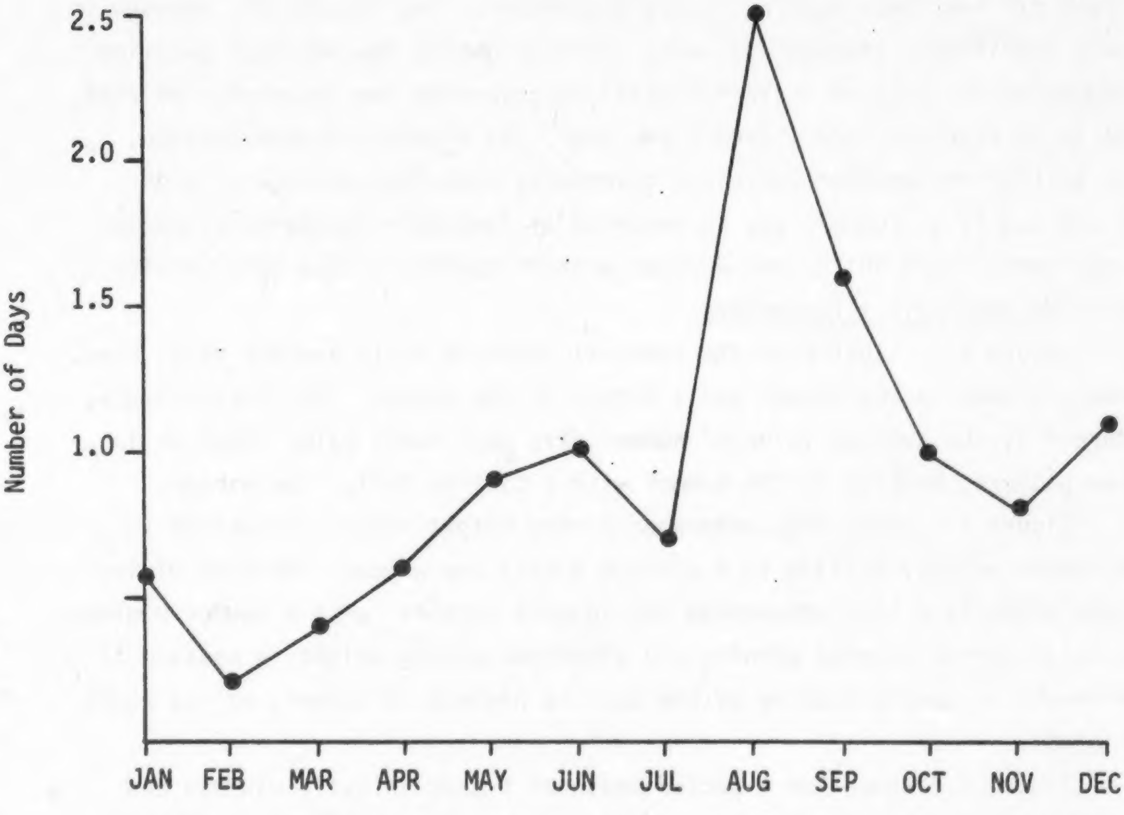

Figure 4.4 Monthly averages of number of days persistent calm (#DY CALM) (Lambert Field data).

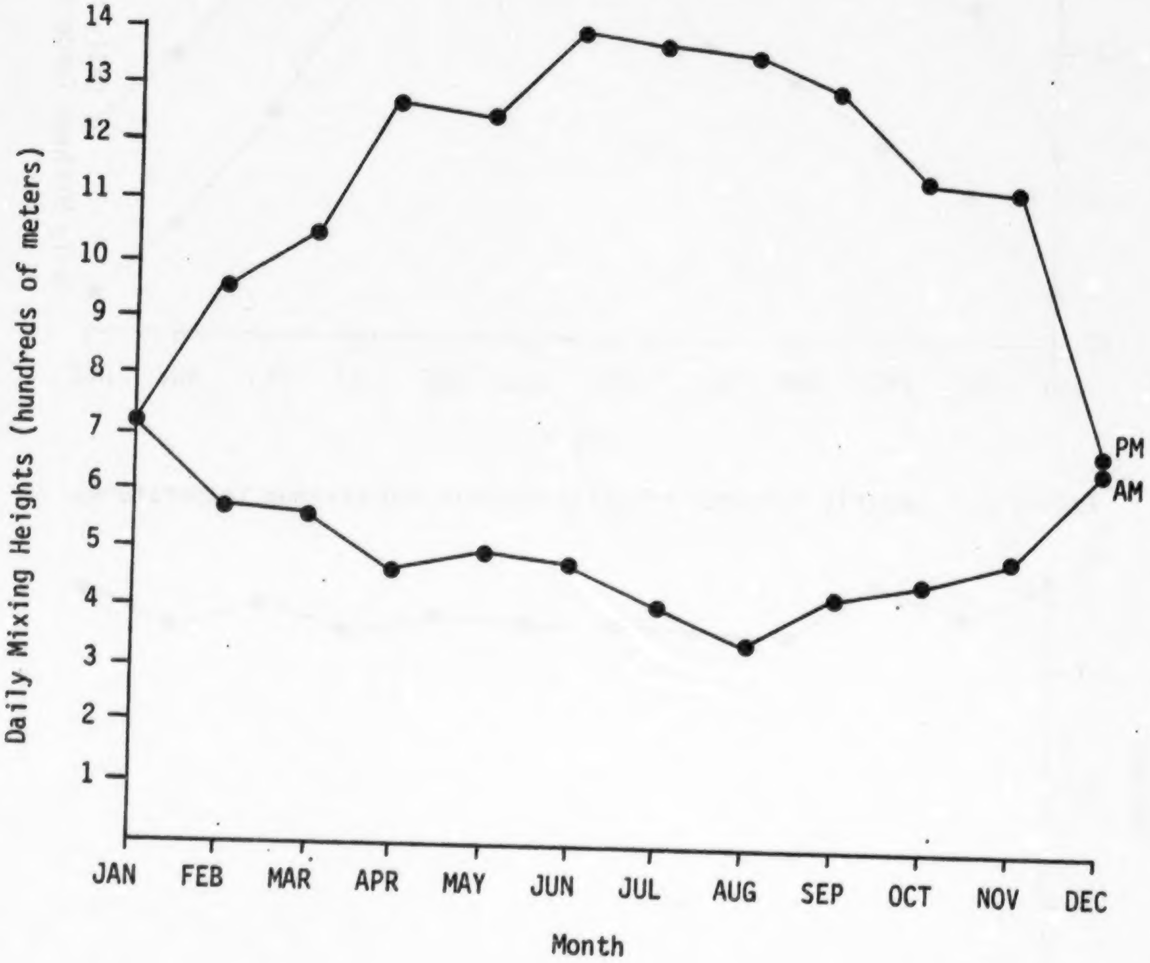

Figure 4.5 Monthly averages of morning and afternoon mixing heights.

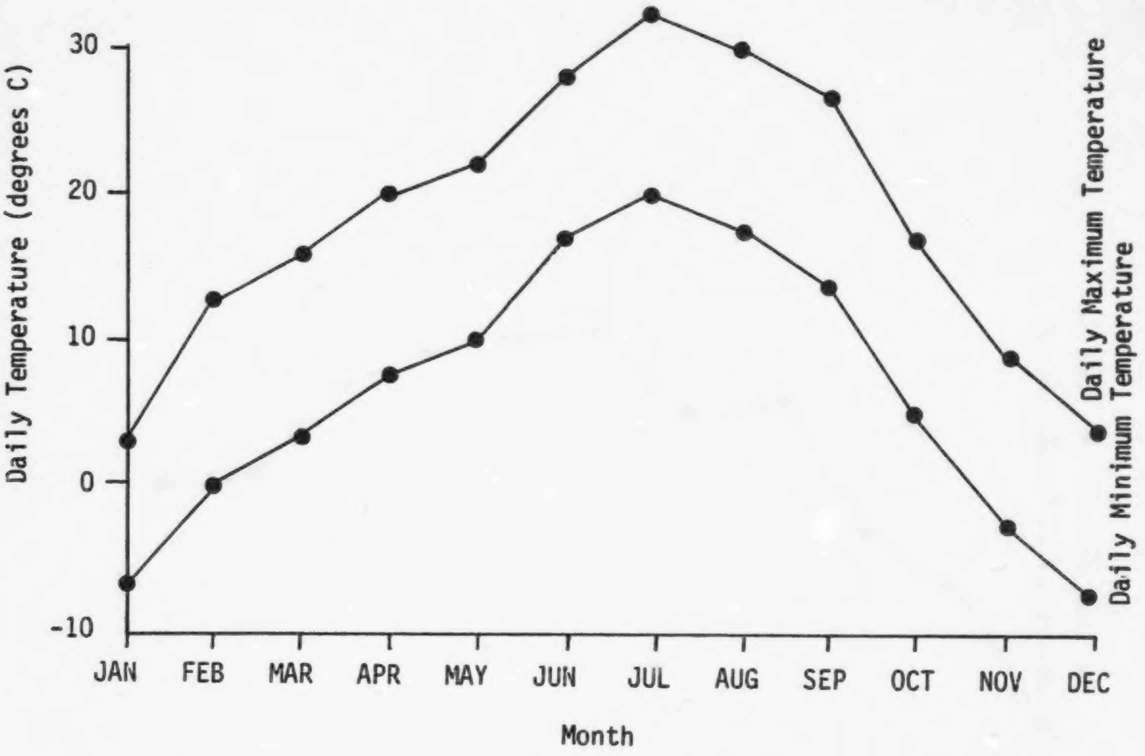

Figure 4.6 Monthly averages of daily minimum and maximum temperatures.

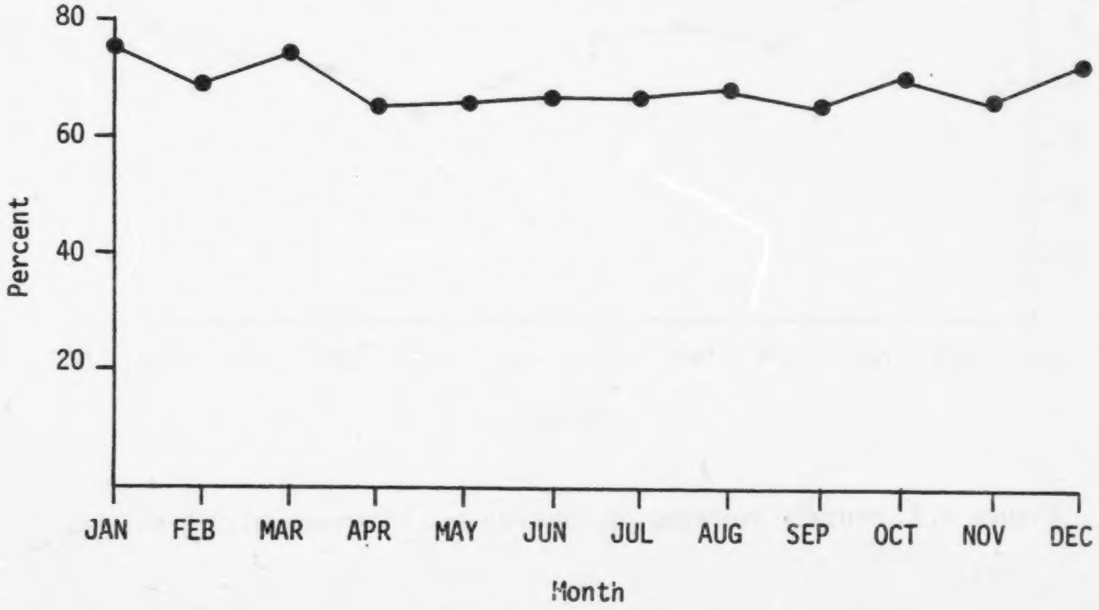

Figure 4.7 Monthly averages of relative humidity.

gap between the minimum and maximum temperature lines is very consistent from month to month.

Figure 4.7 indicates that relative humidity undergoes only a slight seasonal variation, with somewhat higher levels in winter that in summer. Figure 4.8 presents the number of precipitation days per month, showing a slight peak in winter and spring. Figure 4.9, the number of days since last precipitation, exhibits peaks in April and November.

4.1.3 Comparison of Particulate and Meteorological Patterns

The relationship between particulate concentrations and meteorology will be discussed at length in Chapter 5. For sake of completeness, however, it is useful to note that certain relationships observed in Chapter 5 are also apparent in the seasonal patterns. For example, Chapter 5 demonstrates that prolonged stagnations (high #DY CALM) and high temperatures are the two weather conditions closely tied to elevated concentrations of sulfur and FINE. The seasonal patterns are consistent with this since all of these variables show a pronounced summer maximum. Also, Chapter 5 demonstrates that elevated levels of COARSE and the crustal elements occur under dry, calm conditions. The seasonal patterns reflect this because COARSE and the crustal elements display a moderate summer peak, while dryness (as measured by the precipitation variables and relative humidity) and calmness (as measured by wind speeds and calm days) also tend to display summer maxima.

4.2 WEEKLY (HEBDOMADAL) PATTERNS

Since human activities tend to follow pronounced weekly cycles, while natural processes do not, it is informative to look for weekly patterns in particulate concentrations. Of particular interest are differences between weekday and weekend concentrations, since anthropogenic emissions are higher on weekdays than on weekends.

4.2.1 Weekly Patterns at Two Selected Sites

Figures 4.10 through 4.20 present the hebdomadal patterns of IP, FINE, COARSE, and the eight elements at two urban sites, 103 and 105. Of the ten

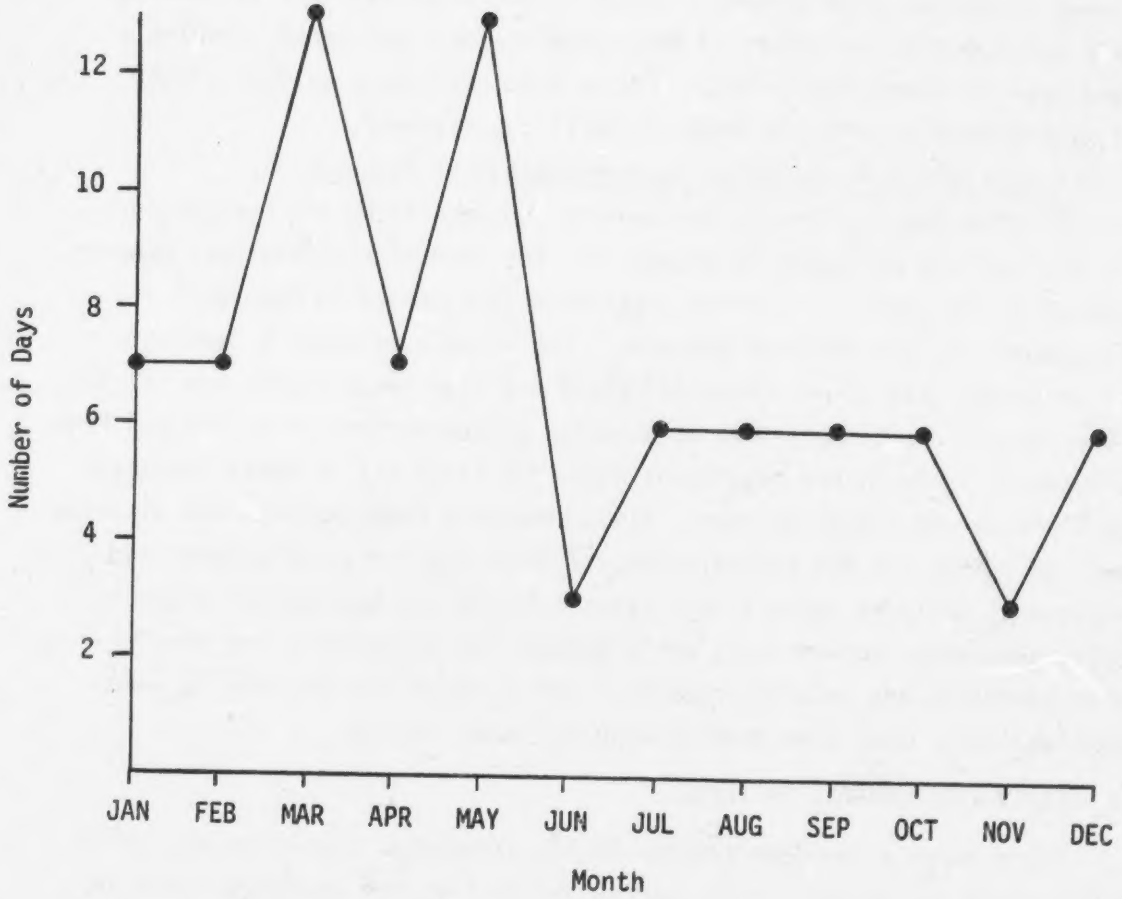

Figure 4.8 Number of days of precipitation per month.

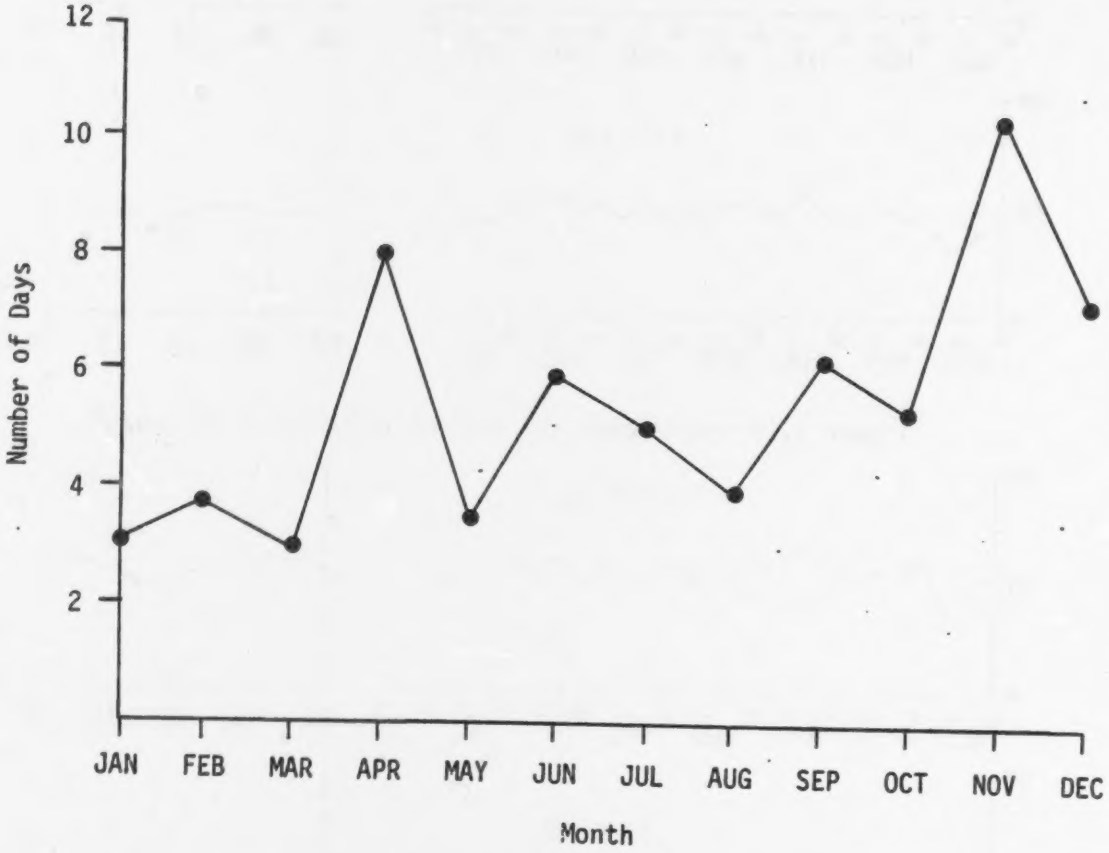

Figure 4.9 Monthly averages of number of days since last precipitation.

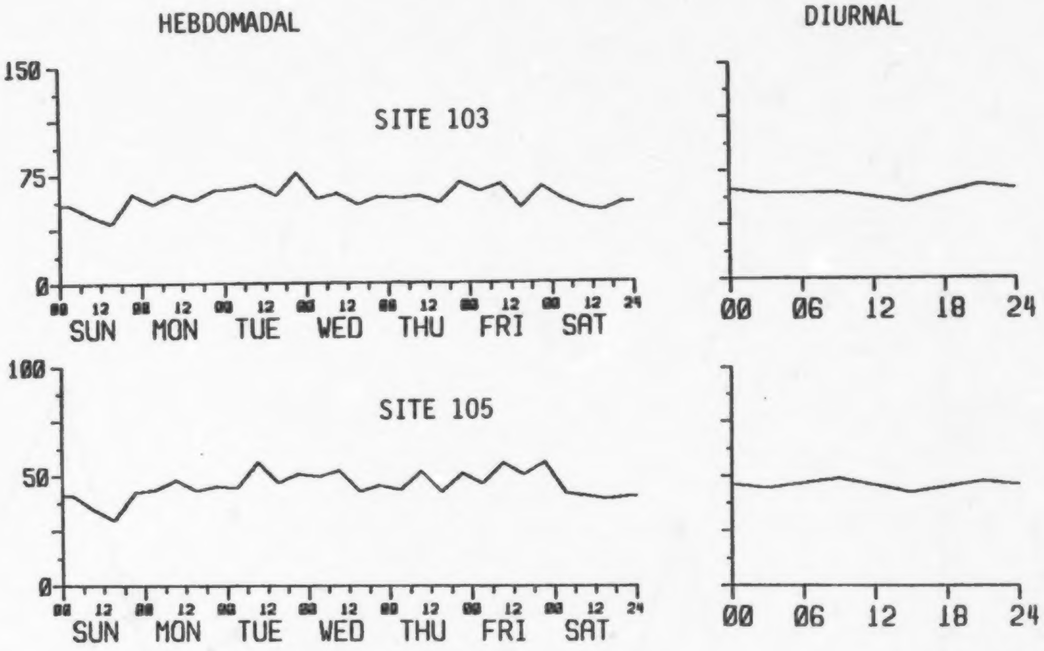

Figure 4.10 Hebdomadal and diurnal patterns of IP ($\mu g/m^3$).

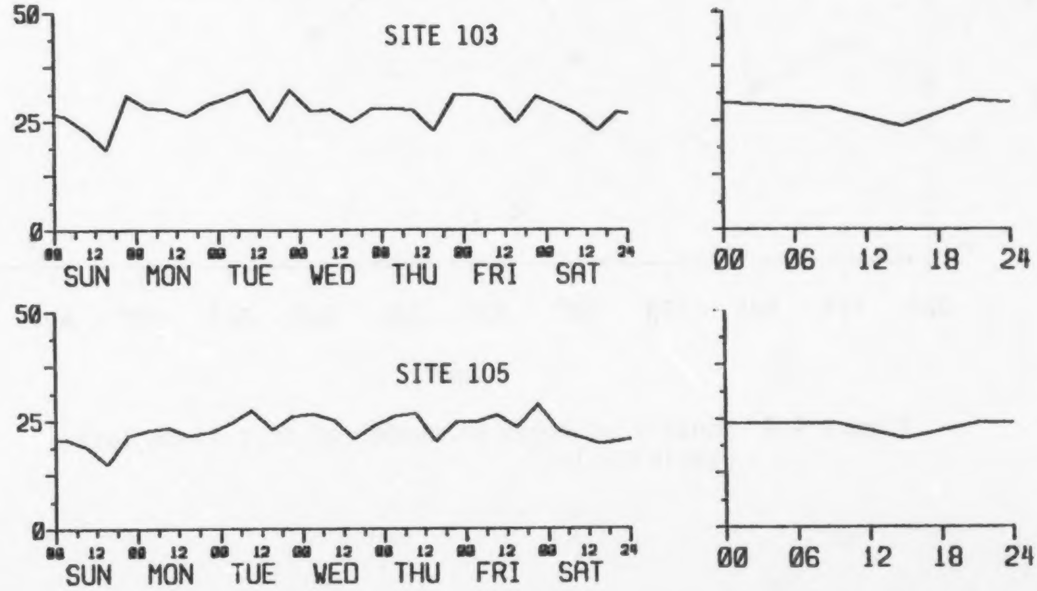

Figure 4.11 Hebdomadal and diurnal patterns of FINE ($\mu g/m^3$).

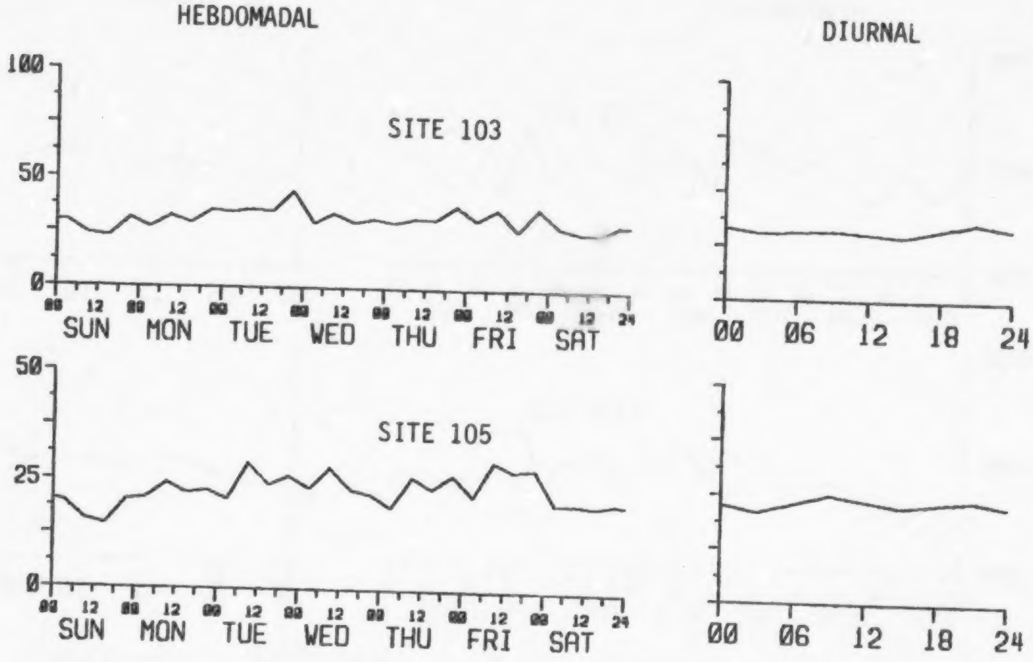

Figure 4.12 Hebdomadal and diurnal patterns of COARSE ($\mu g/m^3$).

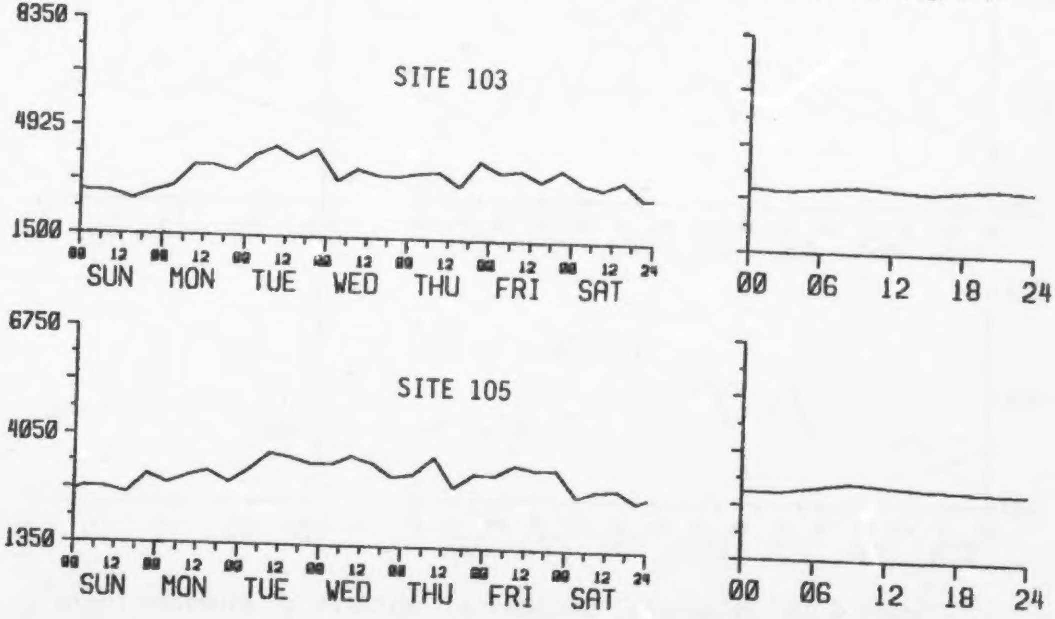

Figure 4.13 Hebdomadal and diurnal patterns of sulfur (ng/m^3).

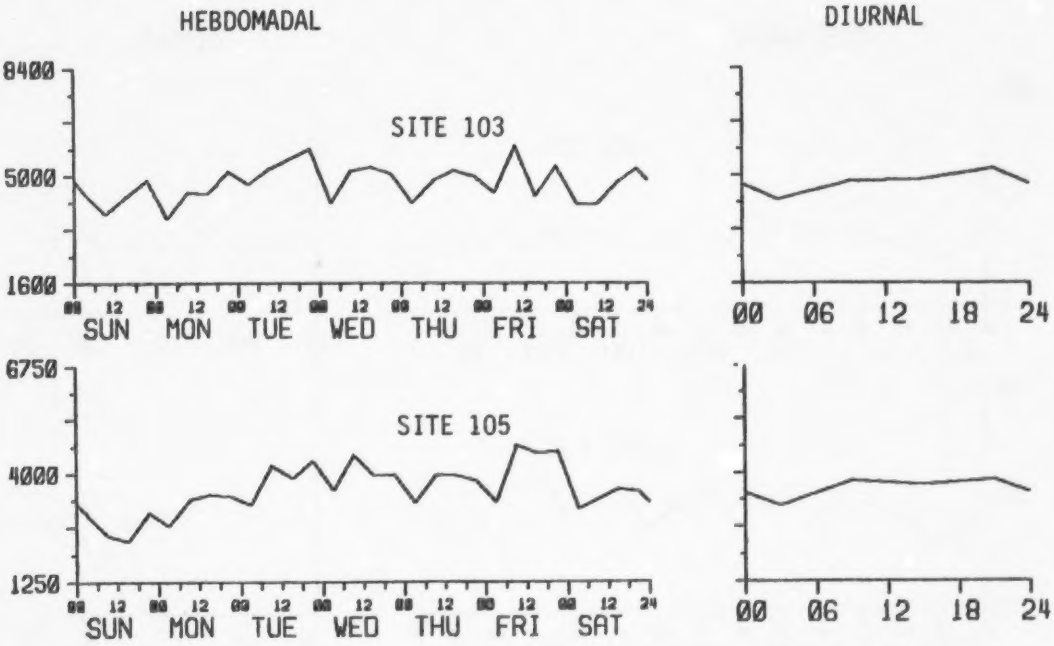

Figure 4.14 Hebdomadal and diurnal patterns of silicon (ng/m^3).

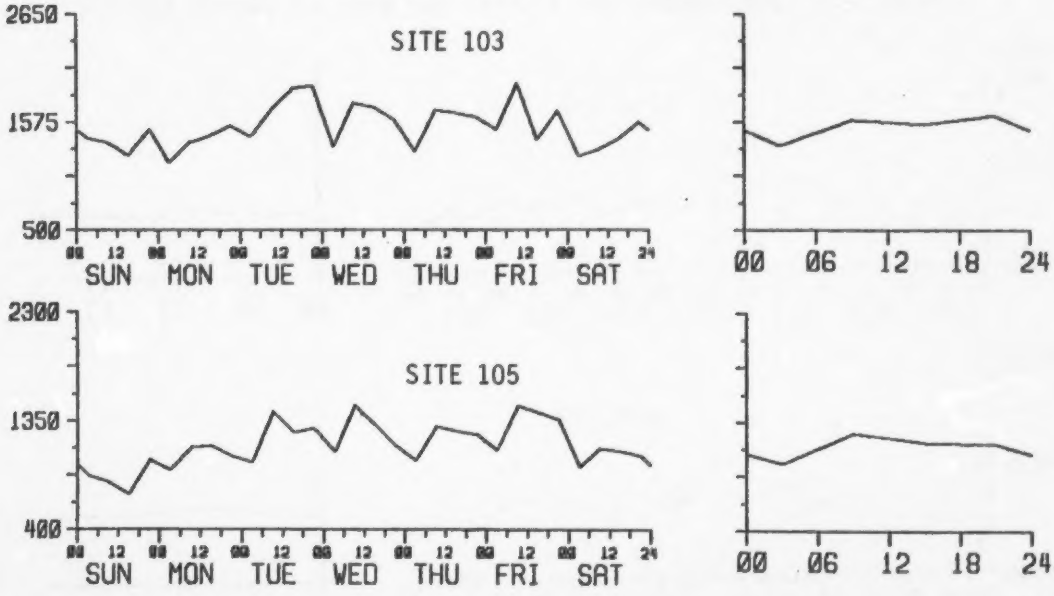

Figure 4.15 Hebdomadal and diurnal patterns of aluminum (ng/m^3).

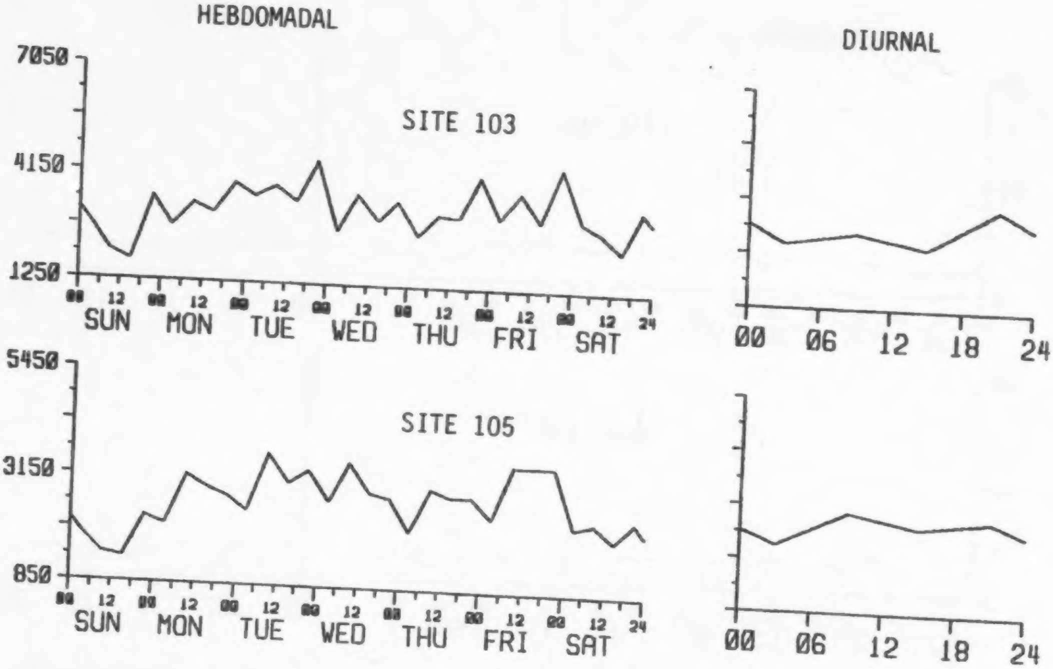

Figure 4.16 Hebdomadal and diurnal patterns of calcium (ng/m^3).

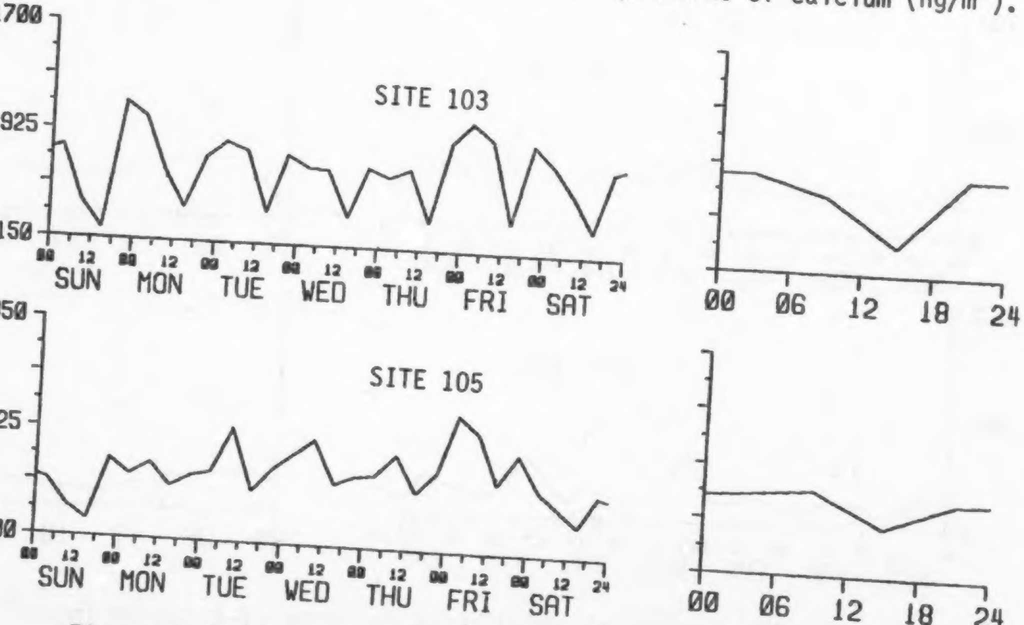

Figure 4.17 Hebdomadal and diurnal patterns of lead (ng/m^3).

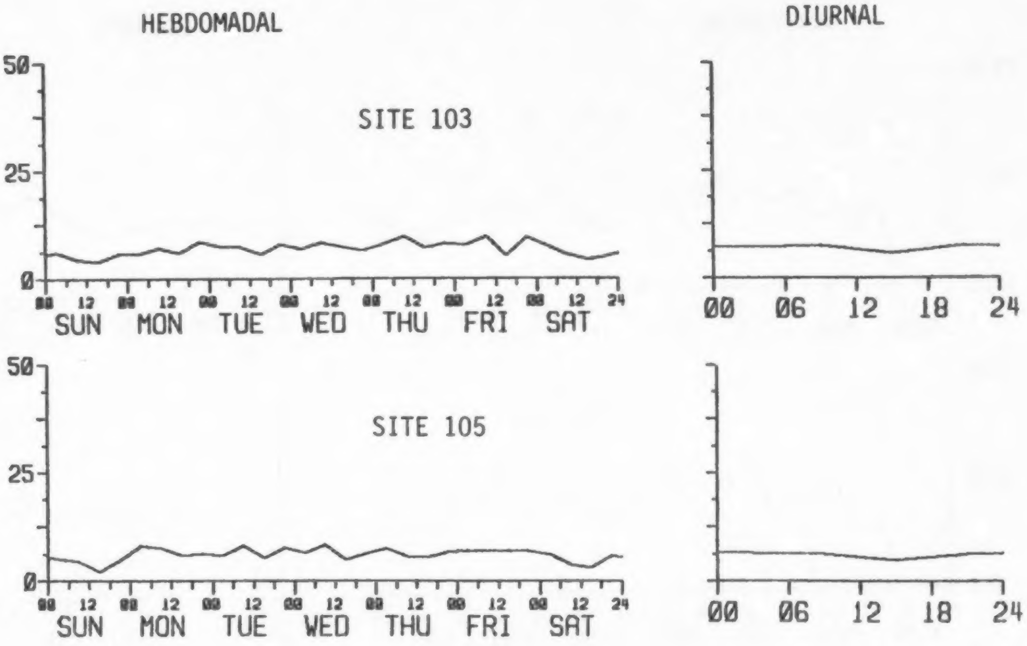

Figure 4.18 Hebdomadal and diurnal patterns of vanadium (ng/m^3).

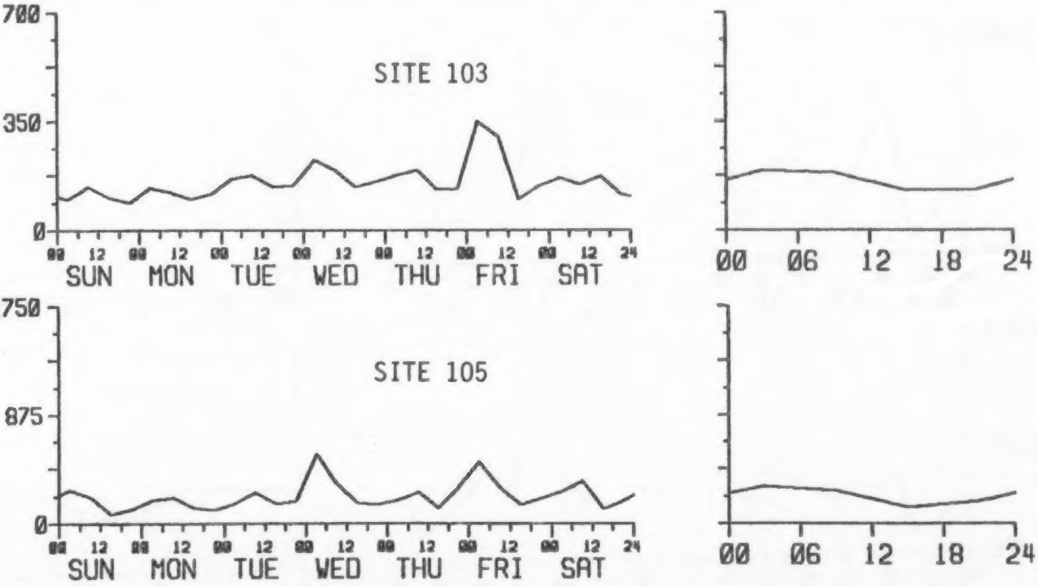

Figure 4.19 Hebdomadal and diurnal patterns of titanium (ng/m^3).

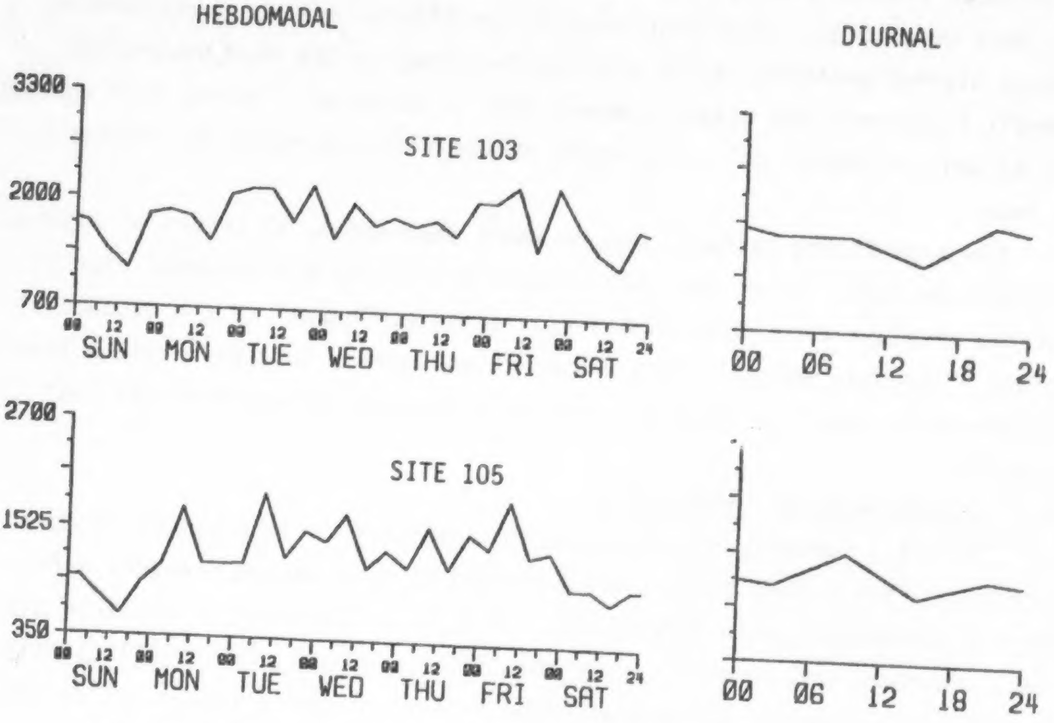

Figure 4.20 Hebdomadal and diurnal patterns of iron (ng/m^3).

dichotomous sampler sites, these two are the only ones at which routine 6-hour data were taken. Also displayed in the figures are the corresponding average diurnal patterns, which will be discussed in the next subsection. Appendix F presents the actual numbers used to construct Figures 4.10 through 4.20 as well as hebdomadal and diurnal patterns disaggregated by quarter of the year.

Few significant patterns emerge among the hebdomadal traces of Figures 4.10 through 4.20. Lead, the only element exhibiting a pronounced diurnal pattern, displays a corresponding series of seven "bumps" in the hebdomadal pattern. The main general feature among the figures, that particulate levels are apparently lower on weekends, will be discussed at length in the next subsection.

4.2.2 Weekend-Weekday Differences

Table 4.1 summarizes weekend-weekday differences in particulate concentrations at the RAMS sites (more detailed data on weekend-weekday differences are presented in Appendix G). Percent differences -- averaged over urban, suburban, and rural sites -- are listed, and the statistical significance of the changes -- as determined by t-tests at individual sites -- are indicated.

Table 4.1 illustrates that the average percent differences, averaged over each type of site, are always negative. Evidently, decreased human activity levels during weekends do lower particulate concentrations. The differences tend to be greatest for the urban sites (especially stations 105, 106, and 108 which are located near the heavily industrialized zones close to the river, towards the southern and northern ends of the city), less for suburban sites, and even less for rural sites. This indicates, as expected, that man-made sources are relatively more important in urban areas than rural areas. The lesser rural weekend-weekday differences may also reflect a shift in the spatial distribution of activity (e.g. traffic); specifically, recreation related activities during the weekends may be relatively more intense in rural areas than are work related activities during weekdays.

TABLE 4.1 WEEKEND-WEEKDAY DIFFERENCES IN ST. LOUIS PARTICULATE CONCENTRATIONS.

PARTICULATE VARIABLE	AVERAGE PERCENT DIFFERENCE[†]			STATISTICAL SIGNIFICANCE									
	Five Urban Sites	Three Suburban Sites	Two Rural Sites	Urban Sites					Suburban Sites			Rural Sites	
				103	105	106	108	112	115	118	120	122	124
TSP	- 9%	- 5%	-12%		**	*			*				
IP	-18%	- 8%	- 8%	*	**	**	**	*					
FINE	-15%	- 7%	-12%		**	**	**						
COARSE	-20%	- 9%	- 4%	*	**	**	**	*					
Sulfur	-18%	-10%	-20%	*	*	*	**					*	*
Silicon	-15%	- 6%	- 4%		**	**	*				*		
Aluminum	-17%	-11%	- 7%		**	**	**				**		
Calcium	-26%	-18%	- 5%	*	**	**	**	*		**	**		
Lead	-22%	-18%	- 1%		**	**		*			**	*	
Vanadium	-29%	-11%	- 5%	*	**	**	**				**		
Titanium	-19%	- 9%	- 3%				*		*				
Iron	-29%	-14%	-10%	*	**	**	**		*		**		

[†] Average percent difference is $\frac{\text{weekend - weekday}}{\text{weekday}}$.

[*] Average weekend concentration is less than average weekday concentration at a 95 percent confidence level.

[**] Average weekend concentration is less than average weekday concentration at a 99 percent confidence level.

The weekend-weekday differences are lower for TSP than for the dichotomous variables. As will be indicated by results in the next chapter, and as makes sense because TSP includes particles in the 20 to 50 μm size range, TSP tends to be more dominated by dust sources than IP. The lower weekend-weekday differences may be a reflection that TSP is also significantly more dominated by wind-blown dust than IP. We cannot be sure of this, however, because the TSP data base is less robust than the IP data base (because of every third day sampling rather than daily sampling); the lesser weekend-weekday differences for TSP may thus be partly a statistical artifact.

As must be, the weekend-weekday differences for IP are intermediate to those for FINE and COARSE. FINE and its most important component, sulfate (or sulfur), are the only particulate variables that show rural weekend-weekday percentage decreases about as large as the urban decreases. This reflects the large spatial scale of sulfates and the other secondary aerosols that constitute the major part of FINE. It is not known how much of the weekend sulfate decrease is related to weekend changes in St. Louis air basin SO_x emissions, larger scale SO_x emissions, and/or photochemical precursor emissions (photochemical smog from hydrocarbons and NO_x promotes more rapid production of sulfate from SO_2). The weekend-weekday differences for sulfur do imply, however, that a significant fraction of sulfate is not related to synoptic scale phenomena. For large scale phenomena, we would expect a considerable time lag between emission changes and air quality changes; this time lag would lessen the likelihood of observing weekend-weekday changes.

The weekend-weekday differences for COARSE are fairly similar to those for the crustal elements (Si, Al, and Ca). This is expected -- other parts of this report indicate that crustal material is the predominant source of COARSE. That significant weekend-weekday differences do exist for the crustal material suggests that most of the crustal material (at least for urban sites) stems from man-made sources; this result is consistent with a major conclusion of Chapter 5 (that the crustal material arises mostly from anthropogenic fugitive dust).

At urban and suburban locations, the weekend-weekday differences for crustal limestone (Ca) are significantly larger than those for crustal shale

(Al and Si). This effect could be explained by the hypothesis that crustal limestone arises relatively more from cement manufacturing and construction (both of which would tend to show very strong weekend changes), while crustal shale arises relatively more from traffic, agriculture, and wind-blown dust. The above hypothesis is also supported by the analysis of spatial patterns (Chapter 2) which showed that Ca has a greater urban/nonurban difference than Si and Al, indicating that Ca is more closely associated with commercial and industrial activities.

The weekend-weekday changes for lead should be fairly representative of patterns in automotive traffic. That these changes are not extremely different from the changes in the crustal elements indicates that traffic may be a significant source of fugitive dust.

Two of the elements that may have significant industrial components, V (fuel oil) and Fe (iron/steel industry), demonstrate the greatest urban weekend-weekday differences. In fact, the large weekend-weekday difference for vanadium implies that a major portion of fuel oil is burned in industrial and commercial sources (residential fuel oil use would show little weekend-weekday change).

4.3 DIURNAL PATTERNS

Figures 4.10 through 4.20 (in the previous section) present diurnal patterns for IP, FINE, COARSE, and the eight elements at sites 103 and 105, the only two sites with routine 6-hour average data. The only parameter showing a pronounced diurnal pattern is lead which demonstrates a distinct afternoon (noon - 6 PM) minimum. This pattern is somewhat puzzling because noon - 6 PM traffic levels should be as great as midnight - 6 AM and 6 PM - midnight traffic levels. An alternative explanation, high afternoon mixing heights, seems contradicted by the fact that none of the other elements exhibit lead's strong diurnal pattern (although Fe and Ti do have a small afternoon dip). Perhaps the explanation is mixing height, with the diurnal patterns for other elements being smoothed by higher source activity in the afternoon or by a complex interaction of diurnal source strength, diurnal mixing height, and particle settling (note that lead, a fine aerosol, would not have the settling effect).

As noted earlier in this report, road dust is suspected of being a significant source of the crustal material that dominates COARSE. Although possibly a significant source of crustal material, road dust appears not to be a predominant source because the diurnal patterns for Al, Si, Ca, and COARSE do not reflect the strong diurnal features exhibited by the automotive tracer, Pb.

5.0 DECISION-TREE ANALYSIS OF PARTICULATE AND METEOROLOGICAL DATA.

One goal of this study is to gain an understanding of the relationship between particulate air quality and meteorology and to see if this understanding can provide insights into the major sources of ambient particulate matter. This chapter investigates the relationship between particulate concentrations and meteorology by using a very general and powerful statistical tool, decision-tree analysis. For three locations (urban sites 103 and 105, and rural site 124), decision-trees are constructed relating each of the twelve particulate parameters (TSP, IP, FINE, COARSE, and the eight elements) to eleven meteorological parameters. Decision-trees are also formed relating IP and FINE to the eight elemental variables.

5.1 THE CART DECISION-TREE PROGRAM

The decision-tree program used in this study is the CART (Classification and Regression Trees) program developed at Technology Service Corporation. The purpose of the program is to explain the variation in a dependent variable (e.g. particulate concentrations) by sequentially splitting the data according to ranges of the independent variables (e.g. meteorological parameters). As illustrated by the example in Figure 5.1, the net result is a decision-tree that accounts for the variance in the dependent variable according to groups (e.g. meteorological classes) defined by the independent variables.

The CART program consists of a rather complicated set of algorithims. The four basic parts of the program are as follows:

I. A set of decision-trees is generated with the entire data base by sequentially splitting the data according to ranges of the independent variables. Each sequential split is performed on the subgroup with the greatest variance; the split is selected to maximize the variance explained, weighted by a factor which favors robust (many data point) splits. The processs is terminated by limits on

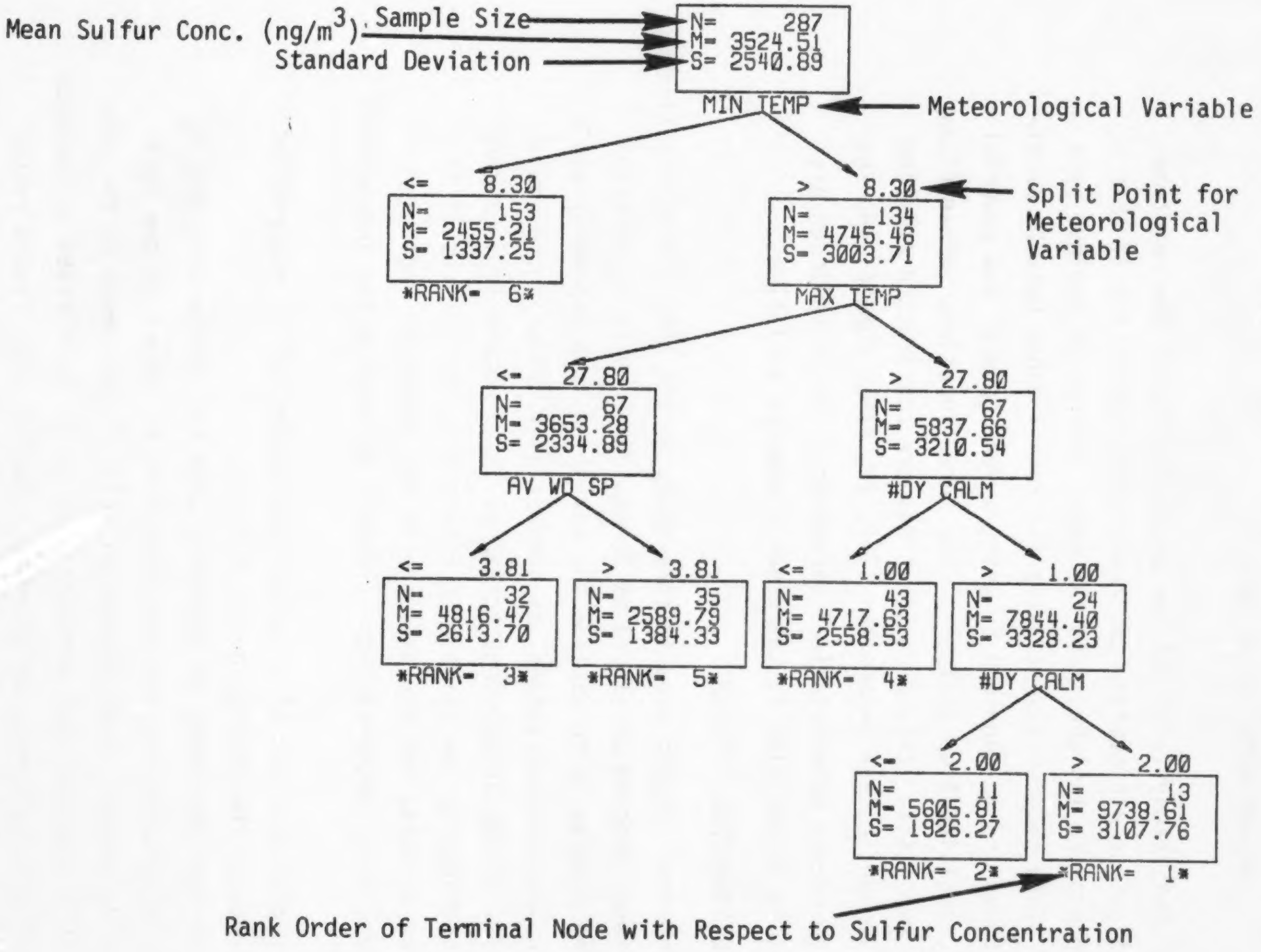
Mean Sulfur Conc. (ng/m3)
Sample Size
Standard Deviation
N= 287
M= 3524.51
S= 2540.89
MIN TEMP
Meteorological Variable
<= 8.30
N= 153
M= 2455.21
S= 1337.25
RANK= 6
> 8.30
Split Point for Meteorological Variable
N= 134
M= 4745.46
S= 3003.71
MAX TEMP
<= 27.80
N= 67
M= 3653.28
S= 2334.89
AV WD SP
> 27.80
N= 67
M= 5837.66
S= 3210.54
#DY CALM
<= 3.81
N= 32
M= 4816.47
S= 2613.70
RANK= 3
> 3.81
N= 35
M= 2589.79
S= 1384.33
RANK= 5
<= 1.00
N= 43
M= 4717.63
S= 2558.53
RANK= 4
> 1.00
N= 24
M= 7844.40
S= 3328.23
#DY CALM
<= 2.00
N= 11
M= 5605.81
S= 1926.27
RANK= 2
> 2.00
N= 13
M= 9738.61
S= 3107.76
RANK= 1
Rank Order of Terminal Node with Respect to Sulfur Concentration

Figure 5.1 Example decision-tree, sulfur at site 103 versus meteorological variables.

the allowable number of terminal nodes and on the minimal allowable improvement in variance explained.[*]

II. The final, most complex decision-tree from part I is subjected to a pruning procedure (a procedure that reduces the complexity of the tree while losing minimal amounts of variance explained). The pruning is carried out sequentially, with the decision-tree at each step graded according to a complexity parameter.

III. The data base is randomly divided into ten parts, each containing 10% of the data. Withholding each of these parts in turn, ten new sets of decision-trees are generated with the remaining ninety percents of the data following the procedures in parts I and II. Each decision-tree in these sets is rated according to percent variance explained using the independent 10% of the data. An optimal complexity parameter is determined by maximizing the average percent variance explained over the ten sets of trees.[**] The pruned tree from part II above (based on all the data) corresponding to the optimal complexity parameter is chosen as the final CART decision-tree. The percent variance explained is taken as the average level achieved by the ten cross-validation tests using independent data sets.

IV. The final aspect of CART allows one to test the importance of each independent variable to the overall decision-tree. For each independent variable, the entire data set is divided into quartiles ranked according to that variable. The variance explained in the predicted values (from the decision-tree) by this quartile split is used as a measure of the importance of the variable to the decision-tree. An example output from this part of the program (corresponding to the decision-tree in Figure 5.1) is presented in Table 5.1.

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[*] The first part of CART is similar to the AID decision-tree program developed at the University of Michigan (Sonquist et al., 1973), except CART involves three refinements: (1) AID requires the independent variables to be specified in discrete form (i.e. continuous variables must be divided into pre-selected ranges), while CART has no such requirement; (2) CART includes special procedures to handle cyclical variables (e.g. wind direction); and (3) AID selects splits solely on percent variance explained, while CART adds a weighting function to favor robust splits.

[**] Note: at a small level of complexity, little variance is usually explained because the data have not been divided enough; at a large level of complexity, loss in variance explained occurs due to over-fitting of the (ninety percent) data sets used to construct the trees.

TABLE 5.1 PERCENT OF VARIANCE IN THE DECISION-TREE FOR SULFUR AT SITE 103 ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES.

METEOROLOGICAL VARIABLE	PERCENT VARIANCE EXPLAINED
#DAYS PP	2%
#DY CALM	16%
%REL HUM	1%
AV WD SP	11%
D7AM PRS	5%
MAX TEMP	27%
MIN TEMP	24%
MIXHT AM	9%
MIXHT PM	13%
PRECIPIT	1%
WIND DIR	6%

For the problem of relating particulate air quality to meteorological variables, the CART decision-tree program offers several advantages over more conventional data-analytic techniques such as multiple linear regression. The advantages include the following:

- The CART program is a non-parametric technique based on a general form of the least-squares principle; it does not involve restrictive assumptions such as additivity and linearity.
- The meteorological classes defined by CART can be interpreted on physical grounds more easily than a multivariate regression equation.

- Because the CART trees are evaluated by a cross-validation test with independent data sets, an unbiased estimate is obtained of the percent variance explained.
- Previous experience (Trijonis et al., 1979), indicates that decision-tree analysis of aerosol/meteorology relationships generally explains more variance than multiple linear regression.

5.2 DECISION-TREES RELATING IP AND FINE TO ELEMENTAL CONCENTRATIONS

Before we proceed to anlayze the decision-trees relating particulate variables to meteorology, it is worthwhile to digress slightly and examine decision-trees relating IP and FINE to elemental concentrations. Because the elemental variables serve as tracers for sources, the relationship of IP (or FINE) to the elements will itself shed light on the major sources contributing to IP (or FINE). Also, knowing how IP (or FINE) depends on the elements will be useful for interpreting the decision-trees relating all the particulate variables to meteorology.

Decision-trees relating IP and FINE to the eight elemental variables at sites 103, 105, and 124 are presented in Appendix H. Table 5.2 summarizes the percent variance explained by the trees; Tables 5.3 and 5.4 summarize the relationships of FINE and IP to the elemental variables.

TABLE 5.2 PERCENT VARIANCE EXPLAINED BY DECISION-TREES RELATING IP AND FINE TO ELEMENTAL VARIABLES.

SITE	PERCENT VARIANCE EXPLAINED	
	IP	FINE
103	71 ± 3	68 ± 3
105	68 ± 3	73 ± 3
124	59 ± 3	68 ± 3

The percent variance explained by the elemental concentrations is quite high, 59% to 71% for IP and 68% to 73% for FINE. These are equivalent to correlation coefficients[*] of 0.77 to 0.84 for IP and 0.83 to 0.85 for FINE.

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[*] Note that the percent variance explained is the square of the correlation coefficient.

TABLE 5.3 RELATIONSHIP OF FINE TO ELEMENTAL CONCENTRATIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL ELEMENTS*							
	S	Si	Al	Ca	Pb	V	Ti	Fe
103	55%	---	---	---	13%	10%	10%	10%
105	56%	15%	17%	16%	---	---	10%	19%
124	55%	---	---	---	---	---	---	---

*Includes only those elements explaining at least 10% of the variance

SITE	CONDITIONS ASSOCIATED WITH HIGH FINE CONCENTRATIONS**
103	high S (6); high Pb (2); high Si (1); high Ti (1).
105	high S (7); high Pb (1); high Ca (1); high Si (1); high Fe (2).
124	high S (5)

**Elements are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this element.

TABLE 5.4 RELATIONSHIP OF IP TO ELEMENTAL CONCENTRATIONS AS FOUND IN THE DECISION-TREE ANALYSIS

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL ELEMENTS*							
	S	Si	Al	Ca	Pb	V	Ti	Fe
103	41%	27%	27%	36%	19%	19%	24%	33%
105	41%	33%	32%	35%	10%	---	23%	42%
124	37%	27%	27%	24%	---	---	29%	28%

*Includes only those elements explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCAITED WITH HIGH IP CONCENTRATIONS**
103	high S (5); high Ca (4); high Si (1); high Fe (2); high V (1); high Al (1).
105	high Fe (4); high S (5); high Si (2); high Ca (1); high Al (3); high V (1).
124	high S (5); high Ca (3); high Si (1), high Fe (1); high Ti (1); low Ti (1).

**Elements are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this element.

The high degrees of percent variance explained are especially notable because they are unbiased (i.e. they have been determined by cross-validation tests with independent data sets).

As indicated by Table 5.3, sulfur is by far the single most important element explaining the variance in FINE. The major reason for this strong relationship is that sulfate aerosol (traced by the element sulfur) alone constitutes approximately one half of FINE (see Chapter 1). Also, the conditions that produce high sulfate may also lead to high concentrations of other fine aerosols, especially other secondary aerosols; this effect would further strengthen the statistical association between sulfate (or sulfur) and FINE. It is notable that, although the other elements do show some link with FINE, none of them consistently bears a strong relationship with FINE. This may imply that the non-sulfate fraction of FINE is a mixture of fairly small contributions from several types of primary and secondary aerosols.

As illustrated by Table 5.4, sulfur and the crustal elements (e.g. Si, Al, and Ca) account for most of the variance in IP. This is because, as noted in Chapter 1, sulfate is the major constituent of FINE while crustal material is the major constituent of COARSE. That the element Fe also is important may be caused by any of three factors: (1) the iron/steel industry may be a significant source of IP, (2) at sites 103, 105, and 124, Fe may represent crustal material rather than the iron/steel industry, or (3) due to intercorrelations in the day-to-day fluctuations of the elements, Fe may be acting as a surrogate for some of the other elements.

5.3 DECISION-TREES RELATING PARTICULATE VARIABLES TO METEOROLOGY

The relationship between ambient particulate concentrations and meteorology can provide insights regarding the sources of particulate matter (Throgmorton and Axetell, 1978; Trijonis et al., 1979; Price et al., 1977). For example, high particulate levels associated with dry conditions and very high wind speed implicates wind-blown dust; high particulate levels associated with dry conditions, high mixing height, and low to moderate wind speed implicates man-made dust sources; high particulate levels associated with low mixing height, low wind speeds, and preferential wind directions tends to imply conventional (e.g. industrial) sources. The reader is referred to Trijonis et al. (1979) for an extensive discussion of the emission source implications that can be drawn from the relationship between aerosol concentrations and meteorological parameters.

This section uses decision-tree analysis to characterize the meteorological conditions associated with high particulate levels and to gain information on sources of ambient particulate matter. Decision-trees are constructed for twelve particulate variables (TSP, IP, FINE, COARSE, and the eight elements) at three RAMS sites (numbers 103, 105, and 124). The eleven meteorological parameters used are summarized in Table 1.2 (Section 1.1). The decision-trees are presented in Appendix I.

Table 5.5 summarizes the percent variance explained by the decision-trees. The results are good to fair for sulfur, FINE, and IP (average percent variance explained on the order of 30%, equivalent to a correlation coefficient of ∿ 0.55); fair to poor for silicon, aluminum, calcium, iron, and COARSE (average percent variance explained on the order of 18%, equivalent to a correlation coefficient of ∿ 0.42); and poor for lead, vanadium, titanium, and TSP (percent variance explained less than 10%).

Despite the less than excellent levels of percent variance explained, the forms of the decision-trees do tend to provide a reasonable and consistent picture of the type of meteorology associated with high particulate levels. Tables 5.6 through 5.17 summarize the particulate/meteorology relationships for the twelve aerosol variables as observed in the decision-trees. The principal conclusions that can be drawn from these tables are as follows:

- Sulfur (Table 5.10)

  Two basic themes stand out in the decision-tree classes for high sulfur: prolonged stagnations (high #DY CALM) and high temperatures. This is consistent with the evidence indicating that particulate sulfur basically represents the secondary aerosol, sulfate (Stevens et al., 1978). The greatest sulfate concentrations are likely to accumulate when calm conditions lead to low dispersal of SO_x emissions, and when intense photochemical activity (due to high temperatures and associated high solar radiation) elevates the sulfate formation rate.

TABLE 5.5 PERCENT VARIANCE EXPLAINED BY DECISION-TREES RELATING PARTICULATE CONCENTRATIONS TO METEOROLOGICAL PARAMETERS.

VARIABLE	PERCENT VARIANCE EXPLAINED*		
	Site 103	Site 105	Site 124
TSP	-2 ± 7	16 ± 6	5 ± 15
FINE	37 ± 6	33 ± 8	34 ± 12
COARSE	25 ± 6	21 ± 8	3 ± 10
IP	37 ± 6	25 ± 10	29 ± 10
Sulfur	31 ± 6	36 ± 7	15 ± 12
Silicon	25 ± 6	15 ± 6	15 ± 12
Aluminum	14 ± 6	26 ± 6	8 ± 8
Calcium	20 ± 6	23 ± 11	17 ± 10
Lead	7 ± 4	-13 ± 17	-25 ± 55
Vanadium	7 ± 5	11 ± 3	10 ± 5
Titanium	-40 ± 57	-4 ± 17	16 ± 6
Iron	39 ± 8	7 ± 5	15 ± 10

[*] The percent variance explained is determined by cross-validation tests with independent data sets and can therefore be negative.

TABLE 5.6 RELATIONSHIP OF TSP TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	----	14%	----	----	11%	----	39%	32%	----	----
105	13%	----	10%	----	----	----	----	41%	37%	----	----
124	12%	----	11%	----	----	19%	21%	22%	27%	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH TSP CONCENTRATIONS**
103	low %REL HUM (1).
105	high #DAYS PP (1).
124	high MAX TEMP (1); low %REL HUM (1); low D7AM PRS (1); high #DAYS PP (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.7 RELATIONSHIP OF IP TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	20%	----	17%	----	26%	19%	21%	24%	----	----
105	----	21%	----	14%	----	21%	15%	23%	22%	----	11%
124	----	14%	----	10%	----	----	----	26%	26%	----	14%

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH IP CONCENTRATIONS**
103	high MAX TEMP (1); low AV WD SP (2); high #DY CALM (1); low %REL HUM.
105	high MAX TEMP (1); low AV WD SP (1); high #DY CALM (2); low MIXHT AM (1); high MIXHT PM (1); high %REL HUM.
124	N to S WIND DIR (1); high MAX TEMP (1); low D7AM PRS (2); high #DAYS PP (1); N to SE WIND DIR (1); low %REL HUM (1); E and SE WIND DIR (1); low AV WD SP (1); high #DY CALM (1); high D7AM PRS (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.8 RELATIONSHIP OF FINE TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	20%	----	14%	----	26%	24%	15%	16%	----	----
105	----	25%	----	16%	----	21%	18%	16%	16%	----	----
124	----	32%	----	----	----	20%	15%	23%	20%	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH FINE CONCENTRATIONS**
103	high MIN TEMP (1); low AV WD SP (2); high MAX TEMP (1); high #DY CALM (3); ESE to S WIND DIR (1).
105	high MIN TEMP (1); NE to S WIND DIR (1); high #DY CALM (3); low AV WD SP (5); S to SW WIND DIR (1); high D7AM PRS (1); low %REL HUM (1).
124	high #DY CALM (4); high MIN TEMP (1); high #DAYS PP (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.9 RELATIONSHIP OF COARSE TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	----	----	14%	----	20%	14%	19%	19%	----	----
105	----	13%	----	----	----	16%	13%	22%	19%	----	----
124	----	----	----	----	----	----	---	13%	12%	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH COARSE CONCENTRATIONS**
103	high MAX TEMP (2); low AV WD SP (3); high #DAYS PP (1).
105	high MAX TEMP (2); high #DY CALM (1); low AV WD SP (3); low MIXHT AM (1).
124	N to S WIND DIR (1); low %REL HUM (2); low MIN TEMP (3); high MAX TEMP (1); low D7AM PRS (1); high #DAYS PP (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.10 RELATIONSHIP OF SULFUR TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	16%	----	11%	----	27%	24%	----	13%	----	----
105	----	25%	----	----	----	25%	24%	10%	11%	----	----
124	----	35%	----	14%	----	12%	----	11%	10%	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH SULFUR CONCENTRATION**
103	high MIN TEMP (1); high MAX TEMP (1); low AV WD SP (1); high #DY CALM (2).
105	high MIN TEMP (2); high #DY CALM (2); high #DAYS PP (1).
124	high #DY CALM (4).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.11 RELATIONSHIP OF SILICON TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	14%	----	28%	----	----	16%	----	16%	17%	----	----
105	10%	----	19%	----	----	18%	13%	20%	21%	----	----
124	----	----	19%	----	----	----	----	----	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH SILICON CONCENTRATION**
103	low %REL HUM (4); high #DAYS PP (2); high MAX TEMP (1); high MIXHT PM (1); low AV WD SP (1).
105	low %REL HUM (1); low D7AM PRS (1); high MAX TEMP (3); high #DAYS PP (1); high MIN TEMP (1); low AV WD SP (1).
124	low %REL HUM (2); low MIN TEMP (1); high #DAYS PP (1); E to S WIND DIR (1); high %REL HUM (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.12 RELATIONSHIP OF ALUMINUM TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	----	19%	----	----	10%	----	14%	18%	----	----
105	14%	----	----	----	----	17%	11%	24%	22%	----	----
124	----	----	12%	----	----	----	12%	10%	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH ALUMINUM CONCENTRATION**
103	low %REL HUM (3); high MAX TEMP (1).
105	high #DAYS PP (2); high MAX TEMP (2); low AV WD SP (1); low %REL HUM (2); high #DY CALM (1); low D7AM PRS (2); high MIN TEMP (1).
124	low MIN TEMP (1); low %REL HUM (2); E to S WIND DIR (1); NE to S WIND DIR (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.13 RELATIONSHIP OF CALCIUM TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	22%	----	16%	----	----	12%	----	25%	19%	----	----
105	13%	12%	----	----	----	11%	----	25%	19%	----	----
124	----	----	----	----	----	----	18%	11%	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH CALCIUM CONCENTRATION**
103	high #DAYS PP (2); low %REL HUM (1); high MAX TEMP (1); low MIN TEMP (1); low AV WD SP (3); high MIXHT PM (1).
105	high #DAYS PP (1); low %REL HUM (2); low AV WD SP (2); NE to S WIND DIR (1); high #DY CALM (2); S and SW WIND DIR (1); low PRECIPIT (1); low MIXHT AM (2), high mixht PM (1).
124	low MIN TEMP (1); high #DAYS PP (1); high MAX TEMP (1); low AV WD SP (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.14 RELATIONSHIP OF LEAD TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	12%	----	13%	----	----	----	10%	----	----	----
105	----	----	----	11%	----	----	----	12%	----	----	----
124	----	----	----	----	----	----	14%	----	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH LEAD CONCENTRATION**
103	low AV WD SP (1).
105	low AV WD SP (2); high #DY CALM (1); high D7AM PRS (1); high MAX TEMP (1); low #DY CALM (1)
124	low MAX TEMP (1); low MIN TEMP (2); SW and W WIND DIR (1); WEST WIND DIR (1); high MIXHT PM (1); high #DAYS PP (1); low D7AM PRS (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.15 RELATIONSHIP OF VANADIUM TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	----	----	----	----	----	----	----	----	----	----
105	----	----	----	----	----	----	10%	----	----	----	----
124	----	----	----	----	----	----	12%	----	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH VANADIUM CONCENTRATION**
103	high MIN TEMP (1); low AV WD SP (1).
105	low MAX TEMP (1); low MIN TEMP (1).
124	low MIN TEMP (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.16 RELATIONSHIP OF TITANIUM TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	----	----	----	----	13%	----	----	----	----	----
105	----	----	----	----	----	----	----	----	----	----	10%
124	----	----	----	----	----	----	----	----	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH TITANIUM CONCENTRATION**
103	high MAX TEMP (2); low %REL HUM (1); low MIN TEMP (2); low AV WD SP (1); low #DY CALM (1).
105	SE to SW WIND DIR (1); low AV WD SP (1); high MAX TEMP (1); high D7AM PRS (1); low MIN TEMP (2).
124	low %REL HUM (1); low MIN TEMP (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

TABLE 5.17 RELATIONSHIP OF IRON TO METEOROLOGICAL CONDITIONS AS FOUND IN THE DECISION-TREE ANALYSIS.

SITE	PERCENT OF VARIANCE IN DECISION-TREE ACCOUNTED FOR BY INDIVIDUAL METEOROLOGICAL VARIABLES*										
	#DAYS PP	#DY CALM	%REL HUM	AV WD SP	D7AM PRS	MAX TEMP	MIN TEMP	MIXHT AM	MIXHT PM	PRECIPIT	WIND DIR
103	----	----	19%	----	----	13%	----	23%	20%	----	----
105	----	12%	----	----	----	13%	----	19%	17%	----	----
124	----	----	----	----	----	----	15%	11%	----	----	----

*Includes only those variables explaining at least 10% of the variance in the decision-tree.

SITE	CONDITIONS ASSOCIATED WITH HIGH IRON CONCENTRATION**
103	low %REL HUM (3); low AV WD SP (3); low MIXHT AM (1); high #DAYS PP (2); high MIXHT PM (1); high MAX TEMP (1); high #DY CALM (1); low #DAYS PP (1).
105	high MAX TEMP (1), high #DY CALM (1).
124	low MIN TEMP (1); high #DAYS PP (1); E to S WIND DIR (1); low %REL HUM (4); E and SE WIND DIR (1); NE to SE WIND DIR (1); low AV WD SP (1); high MAX TEMP (1); low MAX TEMP (1); low D7AM PRS (1).

**Meteorological parameters are listed in approximate order of appearance in decision-tree. Number in parentheses represents the number of splits in the decision-tree made on this parameter.

- FINE (Table 5.8)

  The two most salient features of the decision-tree for FINE, high #DY CALM and high temperatures, are the same as for sulfur. This is consistent with earlier findings (e.g. Chapter 1) that sulfate comprises approximately half of FINE. Prolonged stagnations and high temperatures would also promote the formation of other secondary aerosols (e.g. nitrates and secondary organics) which likely contribute to FINE. A condition that is important to high FINE at sites 103 and 105 -- low average wind speed -- may reflect localized contributions of primary (or possibly secondary) fine aerosols from industrial sources and fuel combustion. The significance of easterly to southerly flows for elevated FINE at sites 103 and 105 may point toward the industrial areas of East St. Louis or, possibly, synoptic sulfate episodes (from the dense SO_x emissions in the Ohio Valley and/or high pressure systems from the Gulf which are known to be conducive to high sulfate). All in all, the decision-trees for FINE implicate man-made sources, especially secondary aerosols, especially sulfates.

- Silicon, Aluminum, Calcium (Tables 5.11, 5.12, and 5.13)

  The overwhelming factor contributing to high levels of the crustal elements is dryness -- low relative humidity, high number of days since precipitation, and low minimum temperature with high maximum temperature (an indication of low relative humidity). The importance of dryness implies that fugitive dust, rather than fly ash, is probably the major source of these elements. A secondary feature of the decision-trees for the crustal elements, especially at the urban sites and especially for calcium, is calm winds (low AV WD SP and high #DY CALM). This probably* implicates anthropogenic sources of dust (e.g. traffic, industries, quarries, construction, etc.) rather than wind blown dust, particularly for the urban sites and particularly for the limestone component.

- COARSE (Table 5.9)

  The meteorological conditions producing high levels of COARSE -- dryness (low %REL HUM, high #DAYS PP, and low MIN/high MAX TEMP) and low wind speed -- are similar to those producing high levels of the crustal elements. This agrees with earlier statements (e.g. Dzubay's results in Chapter 1) that crustal material is the predominant concontributor to COARSE. As noted in the above paragraph, the suspended crustal material appears to be mostly fugitive dust rather than fly ash. Low wind speeds are more important to COARSE than they are to the crustal elements, apparently reflecting the fact that other man-made sources (in addition to the crustal component) make some contribution to COARSE.

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*Another explanation for the negative dependence of crustal elements on wind speed involves a sampling problem. Specifically, the collection efficiency of particles near the upper size cut-off (∿20 μm) tends to decrease with increasing wind speed.

- IP (Table 5.7)
  As expected, the meteorological conditions associated with high IP appear to be a mixture of the conditions associated with high FINE and high COARSE. High temperatures and calm conditions reflect the contributions from FINE (and sulfates). Dry conditions and low wind speeds reflect the contributions from COARSE (and crustal materials).

- TSP (Table 5.6)
  The decision-tree classes for high TSP point toward one major factor -- dryness -- indicating that TSP tends to be dominated by fugitive dust. We would expect TSP to contain relatively more dust than IP because TSP includes larger particles (i.e. particles in the 20 μm to 50 μm size range as well as those below 20 μm).

- Other Elements: Pb, V, T, and Fe (Tables 5.14 to 5.17)
  The decision-trees for the last four elements point toward the specific sources associated with these elements, but these decision-trees are not important in explaining IP or TSP because none of these sources appear to be major contributors to IP or TSP. The decision-tree classes for high lead (a tracer for automotive exhaust) show poor dispersion as the major theme. The one prevalent feature in the decision-trees for vanadium (a tracer for fuel oil) is low temperature; this is expected because fuel oil use for space heating is a winter phenomenon and because dispersion of primary contaminants is often poor in the winter due to strong nocturnal inversions. The decision-trees for iron and titanium appear rather similar to those for the crustal elements (Si, Al, and Ca); dryness and calm winds are the major features. This seems reasonable because Fe and Ti also serve partly as tracers for crustal materials. Calm conditions are more important for Fe and Ti, however, because these elements also serve as important tracers for industrial sources, iron/steel facilities, and a paint pigment plant.

6.0 RELATIONSHIP BETWEEN TSP AND IP

The existing National Ambient Air Quality Standards (NAAQS) for particulate matter pertain to total suspended particulate mass (TSP) as measured by the Hi-Vol sampler. Because of compliance monitoring for the NAAQS, and because of the widespread use of Hi-Vol samplers even before promulgation of the NAAQS, there is a wealth of historical data on ambient TSP concentrations. In the future, EPA may revise the NAAQS by establishing a size-specific particulate standard for inhalable particulate matter (IP) and possibly fine particulate matter (FINE) as measured by the dichotomous sampler (Miller et al., 1979). If this is done, there will be a need to understand the relationship between TSP and IP so that the historical Hi-Vol data can be viewed in better perspecitve relative to the new dichotomous data. This chapter provides some information on the relationship between TSP and IP by analyzing the simultaneous Hi-Vol and dichotomous data at the ten RAMS particulate monitoring sites.

6.1 SCATTERPLOTS OF DICHOTOMOUS DATA VERSUS HI-VOL DATA

One way of investigating the relationship between dichotomous and Hi-Vol measurements is to compare day-to-day variations in the respective data at individual monitoring sites. This can be done by preparing scatterplots and computing least-squares regression lines relating dichotomous variables to Hi-Vol mass at each of the RAMS locations.

Table 6.1 summarizes the results of regression/correlation analyses relating IP, FINE, and COARSE to TSP. Although the correlations for each variable are statistically significant at all locations, TSP explains (on the average among sites) only 47%, 50%, and 24% of the daily variation in IP, COARSE, and FINE, respectively. The lack of an extremely close association between TSP and the dichotomous variables can also be illustrated by scatter plots. Figures 6.1, 6.2, and 6.3 present such plots for sites 108 (typical correlation), 115 (best-case correlation), and 124 (worst-case correlation), respectively. It is obvious from the above analysis that TSP measurements alone cannot adequately explain the day-to-day fluctuations in IP, COARSE, and especially FINE.

TABLE 6.1 CORRELATION/REGRESSION ANALYSIS BETWEEN DAILY VALUES OF DICHOTOMOUS AND HI-VOL DATA.

Table 6.1a IP Versus TSP

STATION	DEGREE OF CORRELATION*		REGRESSION LINE	
	Correlation Coefficient R	Percent of Variance Explained R^2	Slope	Intercept ($\mu g/m^3$)
103	.65	42%	0.49	11.1
105	.67	45%	0.51	-1.5
106	.69	48%	0.57	6.2
108	.65	42%	0.43	11.0
112	.84	70%	0.57	2.3
115	.87	76%	0.74	-4.6
118	.45	20%	0.27	17.1
120	.55	30%	0.47	11.7
122	.88	78%	0.68	-3.4
124	.42	17%	0.20	18.3
Average	.67	47%	0.49	6.8

Table 6.1b COARSE Versus TSP

STATION	DEGREE OF CORRELATION*		REGRESSION LINE	
	Correlation Coefficient R	Percent of Variance Explained R^2	Slope	Intercept ($\mu g/m^3$)
103	.72	52%	0.33	-0.4
105	.77	59%	0.29	-3.8
106	.71	51%	0.29	1.6
108	.70	49%	0.28	1.5
112	.86	75%	0.37	-5.1
115	.89	79%	0.44	-6.2
118	.44	20%	0.14	7.2
120	.57	33%	0.26	3.0
122	.89	79%	0.40	-5.1
124	.30	9%	0.09	7.6
Average	.69	50%	0.29	.03

Table 6.1c FINE Versus TSP

STATION	DEGREE OF CORRELATION*		REGRESSION LINE	
	Correlation Coefficient R	Percent of Variance Explained R^2	Slope	Intercept ($\mu g/m^3$)
103	.41	17%	0.17	10.4
105	.51	26%	0.22	2.9
106	.55	30%	0.25	5.9
108	.53	28%	0.16	8.8
112	.59	35%	0.21	6.4
115	.57	33%	0.26	4.0
118	.31	9%	0.12	9.9
120	.29	8%	0.14	11.5
122	.62	39%	0.25	3.7
124	.33	11%	0.11	10.7
Average	.47	24%	0.19	7.4

*Note that all correlation coefficients are statistically significant at 95% confidence level.

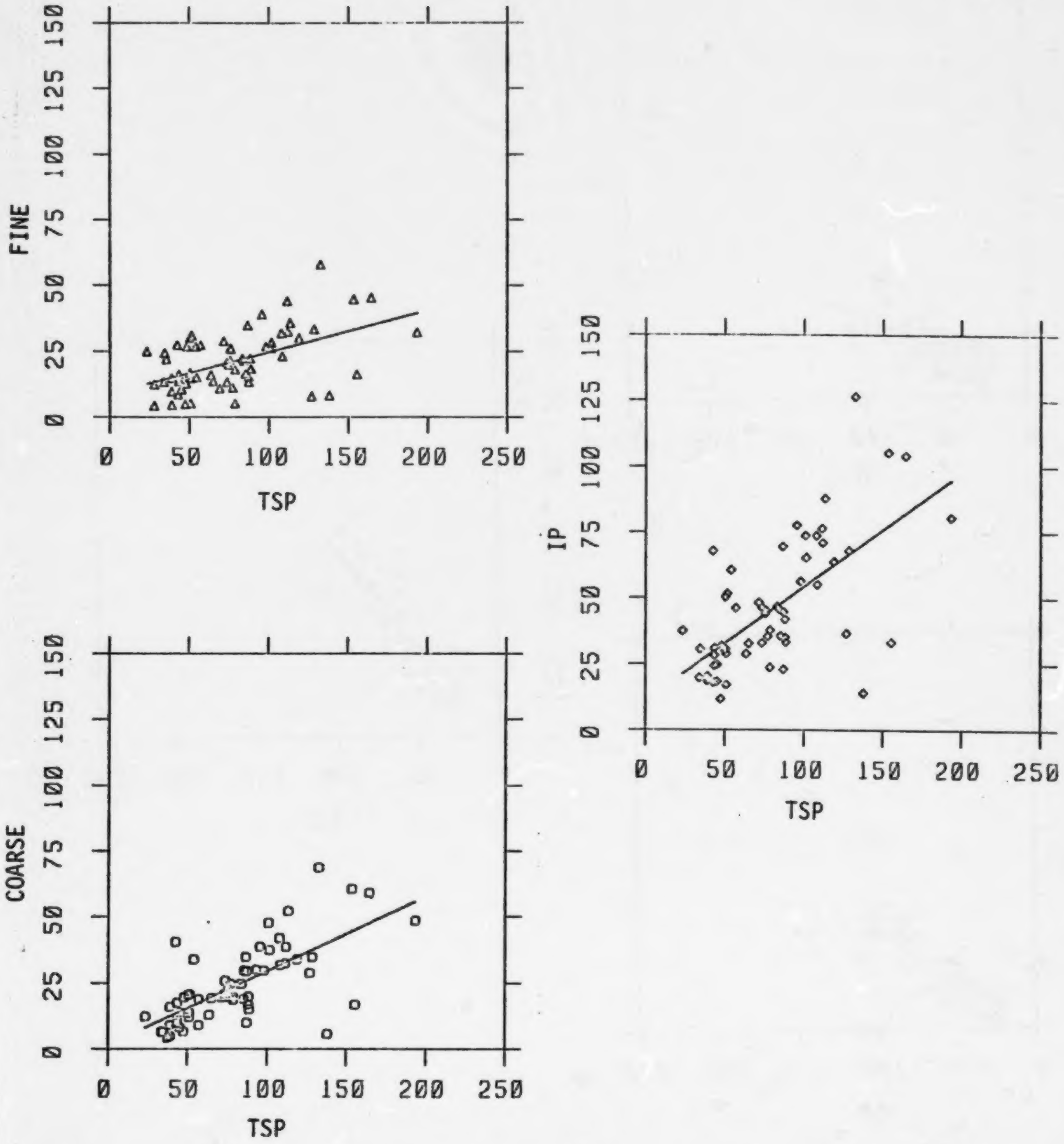

Figure 6.1 Scatterplot of dichotomous variables versus TSP at site 108.

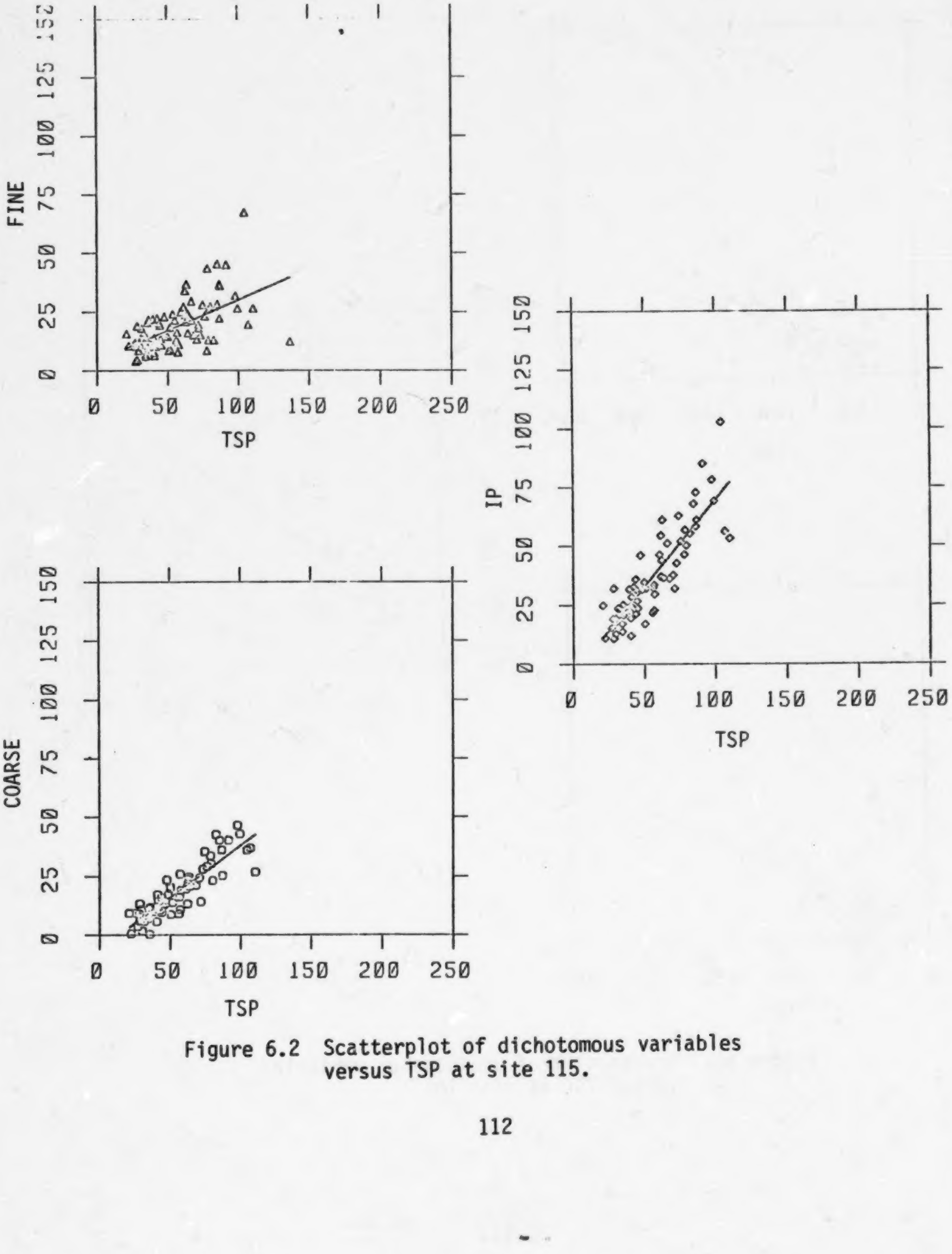

Figure 6.2 Scatterplot of dichotomous variables versus TSP at site 115.

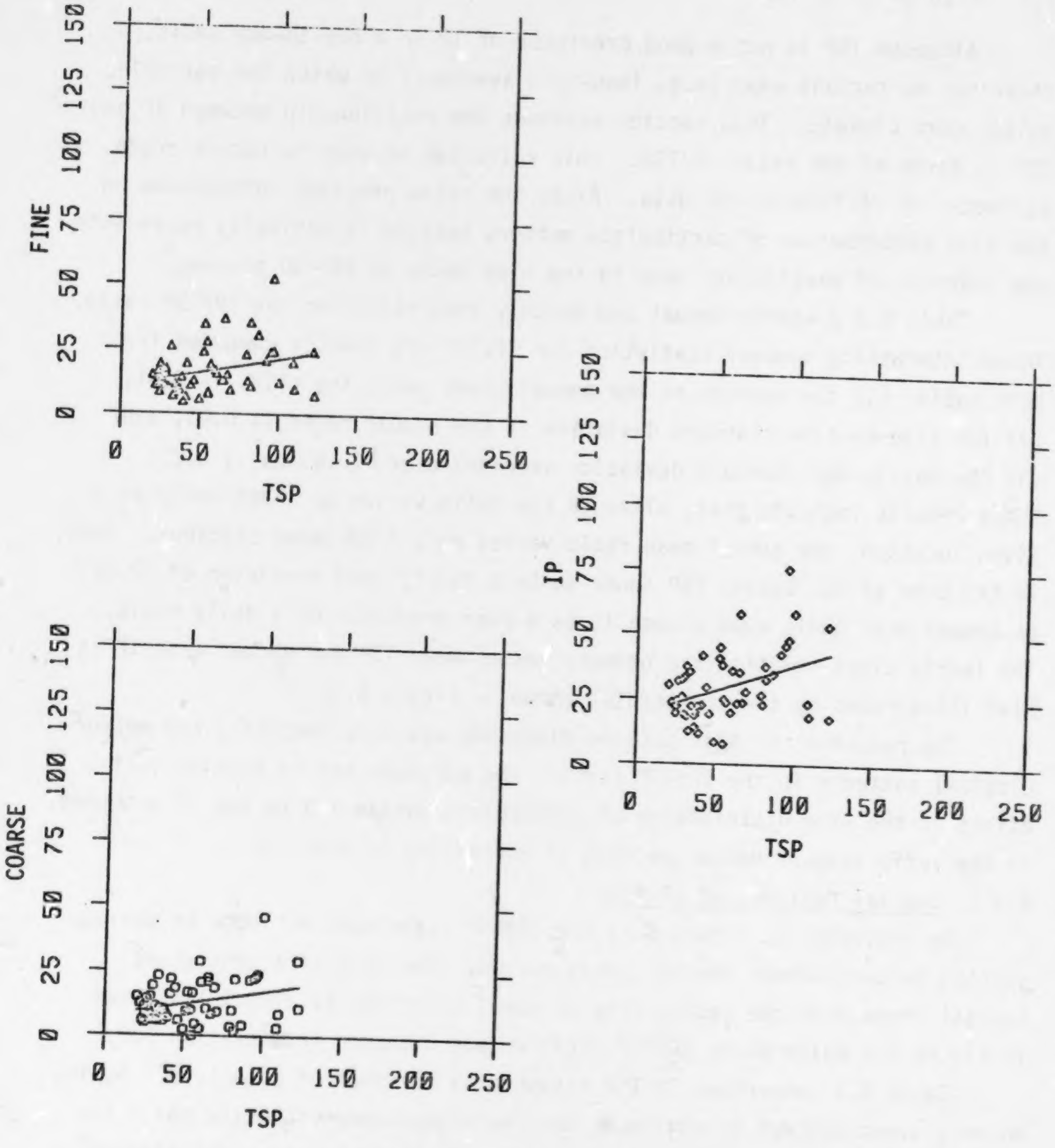

Figure 6.3 Scatterplot of dichotomous variables versus TSP at site 124.

6.2 RATIO OF IP TO TSP

Although TSP is not a good predictor of IP in a day-to-day basis, there may be certain ways (e.g. long-term averages) in which the variables relate more closely. This section examines the relationship between IP and TSP in terms of the ratio IP/TSP. This ratio can be used to obtain crude estimates of IP from Hi-Vol data. Also, the ratio provides information on the size distribution of particulate matter, because it basically represents the fraction of particulate mass in the size range of 20 - 50 microns.*

Table 6.2 presents annual and monthly statistics for the IP/TSP ratio. Three interesting summary statistics for IP/TSP are readily computed from this table: (1) the average of the annual means among the sites is 0.61, (2) the site-to-site standard deviation of the annual means is 0.06, and (3) the day-to-day standard deviation averaged over all sites is 0.22. These results indicate that, although the ratio varies by ± 36% daily at a given location, the annual mean ratio varies only ± 10% among stations. Thus, in the case of St. Louis, TSP tends to be a fairly good predictor of IP on an annual mean basis even though it is a poor predictor on a daily basis. The fairly close relationship between annual mean TSP and annual mean IP is also illustrated by the scatterplot shown in Figure 6.4.

The remainder of this section discusses spatial, temporal, and meteorological patterns in the IP/TSP ratio. The purposes are to examine variations in the size distribution of particulate matter and to see if patterns in the ratio suggest better methods of estimating IP from TSP.

6.2.1 Spatial Patterns of IP/TSP

As indicated by Figure 6.5, the IP/TSP ratio does not show an obvious spatial pattern within the St. Louis region. The lack of a consistent spatial trend from the center city to rural locations is also illustrated in Figure 6.6 which plots IP/TSP ratio versus distance from station 103.

Table 6.3 summarizes IP/TSP ratios as a function of site type. Again, no consistent pattern is obvious. The one urban/commercial site has a low ratio, and the one suburban/industrial site has a high ratio; definitive conclusions, however, cannot be reached based on data from single locations.

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*It does not exactly represent the mass in the size range less than 20 - 50 μm because of particle size - sampling efficiency considerations with both the dichotomous and Hi-Vol samplers, and because of artifact aerosol formation on Hi-Vol filters.

TABLE 6.2 ANNUAL AND MONTHLY STATISTICS FOR IP/TSP RATIO.

STATION	103	105	106	108	112	115	118	120	122	124
NUMBER	73	84	56	60	75	67	73	63	65	53
ARITH MEAN	.63	.49	.64	.60	.61	.65	.55	.71	.61	.61
ARITH SD	.23	.20	.18	.27	.20	.18	.23	.25	.17	.31
MAXIMUM	2.07	1.11	1.18	1.60	1.28	1.18	1.29	1.46	1.02	1.46
2ND HIGHEST	1.18	1.05	.99	1.53	1.16	1.11	1.19	1.33	1.01	1.21
MINIMUM	.23	.20	.14	.10	.27	.30	.11	.18	.16	.14
JAN NUMBER	7	10	9	8	6	9	5	3	9	11
ARITH MEAN	.44	.34	.50	.60	.62	.68☆☆	.49	.52	.52	.96☆
FEB	7	5	5	8	1	7	4	7	4	5
	.44	.45	.54	.81☆☆	.48	.62	.39	.53	.64	.49
MAR	9	10	8	5	9	6	9	9	7	7
	.63	.41	.53	.49	.57	.64	.43	.69	.51	.39
APR	6	9	6	8	9	5	9	7	6	9
	.62	.44	.53	.43	.48	.61	.52☆	.70	.45	.38
MAY	5	3	6	8	8	8	3	9	7	5
	.63	.75☆	.67	.63	.57	.59	.60	.76	.68	.66
JUN	6	10	7	4	9	3	4	3	7	7
	1.15☆	.81☆☆	.83☆	.37	.82☆	.92	.73	.83	.84☆☆	.56
JUL	6	7	6	1	5	6	3	4	3	5
	.65	.43	.75	.31	.74	.75	.59	.97	.67	.47
AUG	6	6	9	2	6		3	6	2	4
	.37	.54	.73	.39	.63		.59	1.07☆	.67	.86
SEP	7	7		5	4	6	9	3	6	
	.69	.51		.53	.46	.63	.61	.74	.60	
OCT	3	7		2	8	3	7	4	2	
	.43	.45		.46	.56☆☆	.53	.42	.63	.97☆	
NOV	4	5		5	6	5	3	3	3	
	.49	.46		.34	.59	.62	.70	.40	.53	
DEC	7	5		4	4	9	7	5	9	
	.53	.37		.79☆	.67	.61☆	.81☆☆	.55	.61	

☆ Month in which maximum occurs.
☆☆ Month in which second highest occurs.
(If the months coincide only one month is flagged.)

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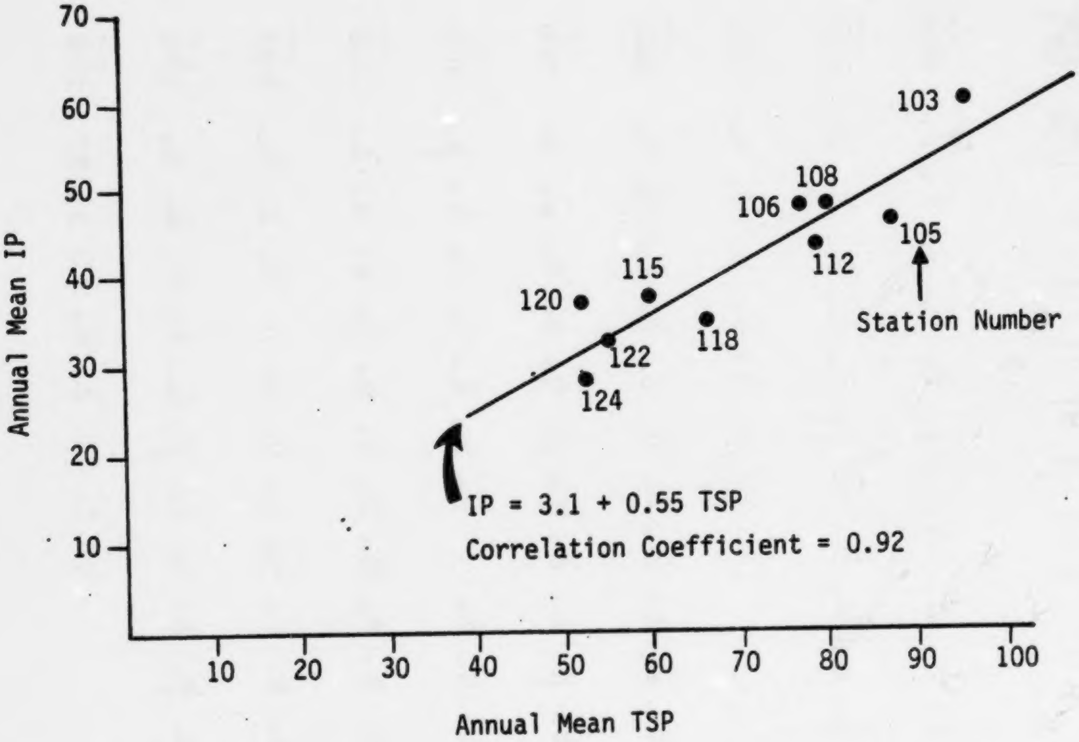

Figure 6.4 Relationship between annual mean IP and annual mean TSP among the RAMS sites.

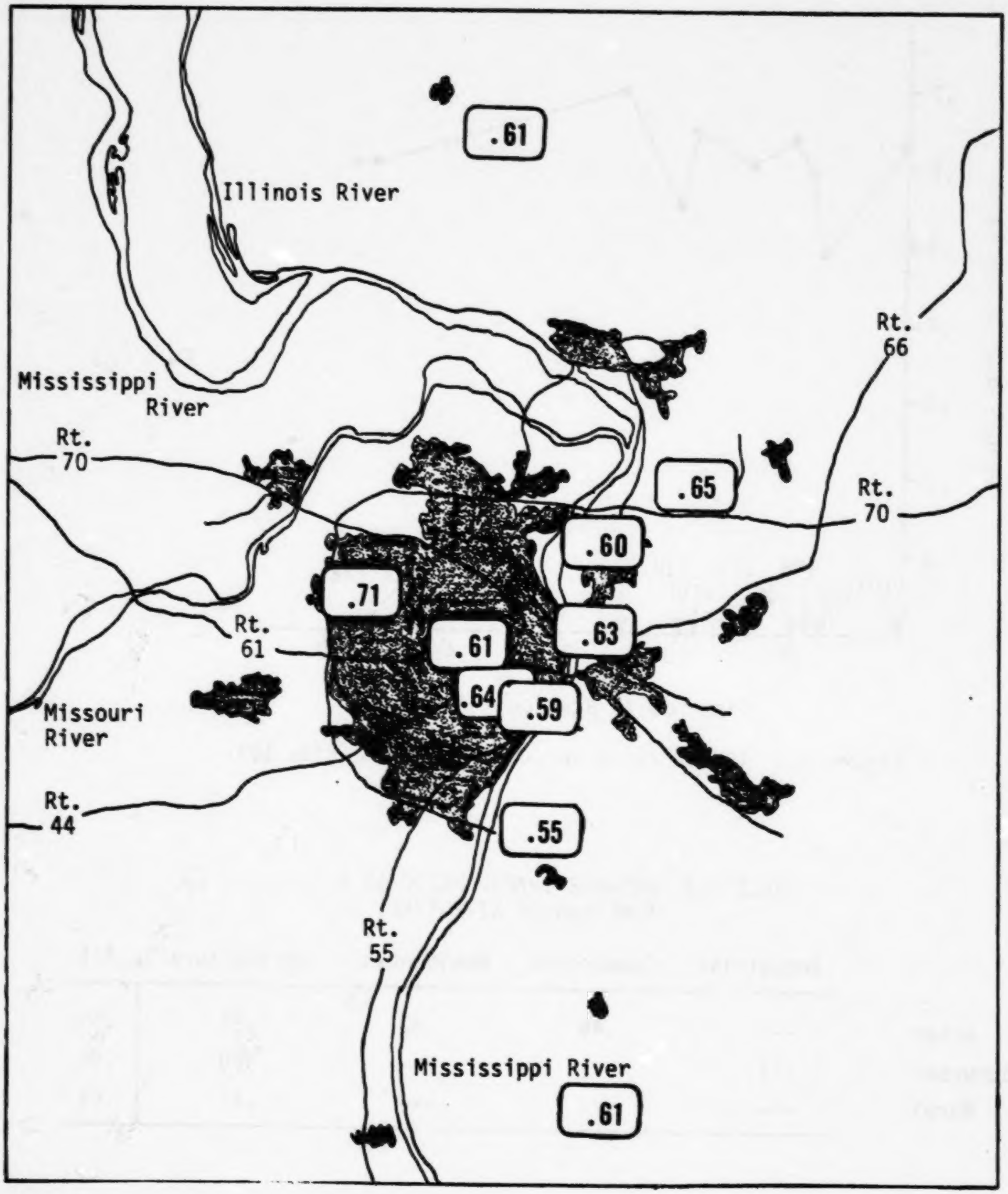
.61
Illinois River
Mississippi River
Rt. 66
Rt. 70
.65
Rt. 70
.60
.71
Rt. 61
.61
.63
.64
.59
Missouri River
Rt. 44
.55
Rt. 55
Mississippi River
.61

Figure 6.5 Geographical distribution of IP/TSP ratio in the St. Louis area.

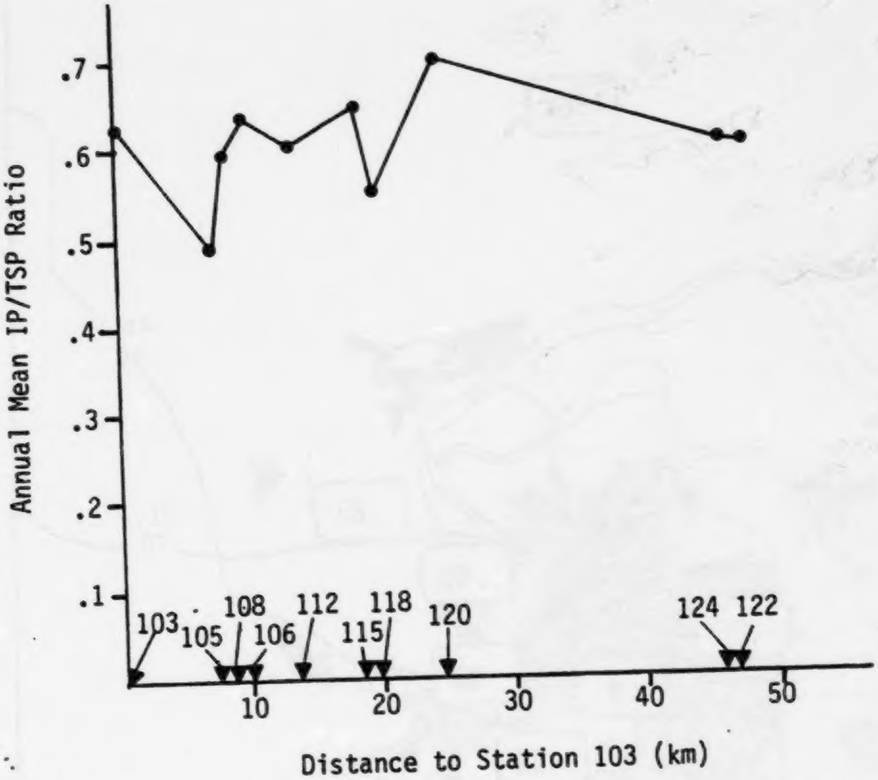

Figure 6.6 IP/TSP ratio versus distance to site 103.

TABLE 6.3 AVERAGE IP/TSP RATIO AS A FUNCTION OF SITE TYPE.

	Industrial	Commercial	Residential	Agricultural	All
Urban	---	.49	.63	.62	.59
Suburban	.71	---	---	.60	.64
Rural	---	---	---	.61	.61

6.2.2 Temporal Patterns of IP/TSP

Figure 6.7 presents the seasonal pattern of the IP/TSP ratio, averaged over all 10 RAMS sites. It can be seen that the IP/TSP ratio tends to be highest in the summer (especially in June), meaning that a greater fraction of the ambient aerosol is less than 20 microns in the summer. Part of this effect is due to the high sulfate concentrations during the summer (see Chapter 4); however, even subtracting out the sulfate influence[*], we have found that a summer peak in the ratio still exists (possibly reflecting photochemical aerosols--other than sulfate--which may also peak during the summer).

Figure 6.8 illustrates the lack of a weekly pattern in the IP/TSP ratio. We have also compared weekend versus weekday levels of the IP/TSP ratio and have found no significant differences.

6.2.3 Decision-Tree Analysis of IP/TSP

In an attempt to gain a better understanding of the factors affecting the IP/TSP ratio on a day-to-day basis, two decision-tree analyses[**] were performed--one relating daily IP/TSP to dichotomous elemental concentrations, and the other relating daily IP/TSP to meteorological parameters. The analyses were conducted for three sites: stations 103, 105, and 124. The resulting decision-trees are presented in Appendix J.

Table 6.4 summarizes the percent variance explained in IP/TSP by the decision-trees[***]. It is obvious that, with the possible exception of the elemental concentration analysis at site 105, the daily variance in IP/TSP is not explained well by either elemental concentrations or meteorological parameters. Despite this lack of success in explaining much of the variance in IP/TSP, the decision-trees did provide some insight into at least one

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[*] To factor out the influence of sulfate on the IP/TSP ratio, we examined the variable (IP - SULFATE)/(TSP - SULFATE), where SULFATE = 4.1 x (dichotomous sulfur).

[**] See Chapter 5 for a discussion of the decision-tree method.

[***] Note that, because the performance of the decision-trees is determined using independent data sets, the percent variance explained can be negative.

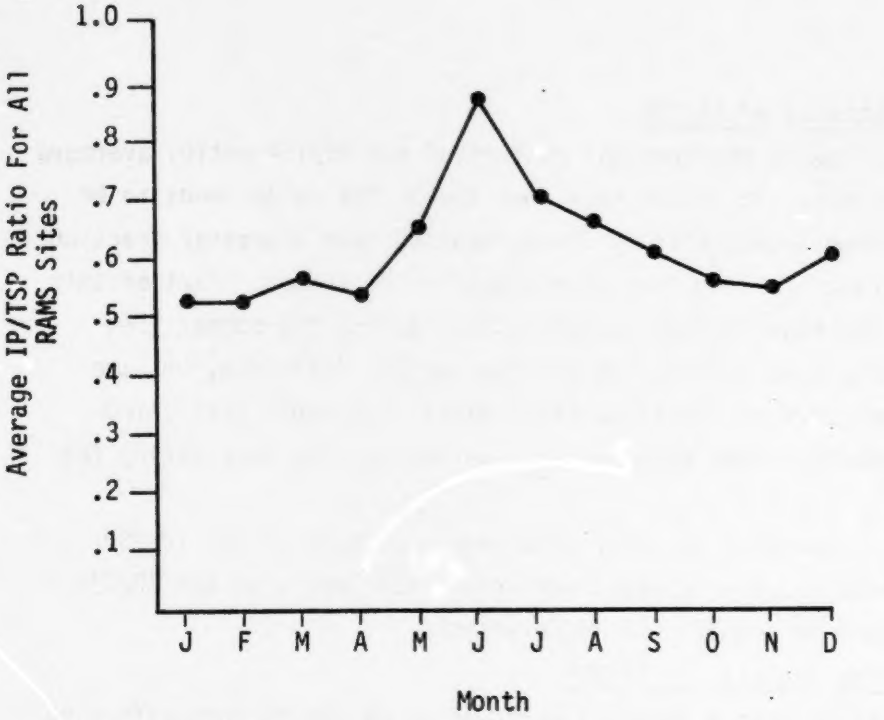

Figure 6.7 Seasonal pattern of the IP/TSP ratio.

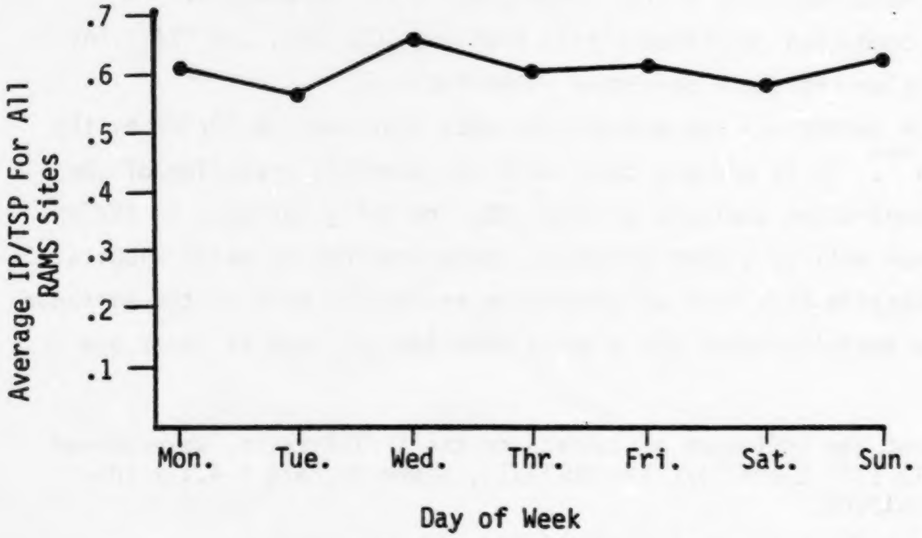

Figure 6.8 Weekly pattern of the IP/TSP ratio.

factor that affects the IP/TSP ratio. Specifically, the elemental decision-trees for sites 103 and 105 indicated that high IP/TSP ratios correlated with high sulfate (sulfur). Also, high IP/TSP ratios correlated with meteorology known or suspected to cause high sulfate[*] (e.g. high temperatures, prolonged stagnation, and high relative humidity). This finding agrees with the results of the seasonal analysis which indicated that elevated levels of sulfate and other secondary aerosols might be associated with the occurrence of high IP/TSP ratios.

TABLE 6.4 PERCENT VARIANCE EXPLAINED IN THE IP/TSP RATIO BY DECISION-TREE ANALYSES.

	PERCENT VARIANCE EXPLAINED[†]	
STATION NUMBER	Decision-Tree Analysis Using 8 Elemental Concentrations	Decision-Tree Analysis Using 11 Meteorological Parameters
103	17% ± 10%	6% ± 12%
105	40% ± 13%	15% ± 10%
124	-28% ± 16%	-31% ± 37%

[†] The reader is referred to Chapter 1 for explicit definitions of the elemental and meteorological variables.

6.2.4 Conclusions Regarding the IP/TSP Ratio

In this section, we have found that the annual mean IP/TSP ratio averaged among RAMS sites is 0.61, with a site-to-site standard deviation of ± 0.06. Also, the day-to-day deviation away from the annual mean at individual stations is ± 0.22. Beyong these rather simple conclusions in regard to the IP/TSP ratio in St. Louis, we have found very little. The geographical patterns, temporal patterns, and decision-tree analyses do not support refined estimates of the ratio with respect to position, time, or meteorology. The only possible exception arises from the seasonal pattern, i.e. the ratio appears to be higher in the summer than in the winter. Even this exception, however, may be in doubt because recent data for Topeka (an urban area in the same region of the country) show the opposite of the seasonal pattern observed in St. Louis (Spengler, 1979).

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[*] See Chapter 5 for a discussion of the analysis relating sulfur concentrations to meteorology.

7.0 SOURCES OF PARTICULATE MATTER IN ST. LOUIS

Evidence concerning the sources of ambient particulate matter is spread throughout this report: Dzubay's chemical element balance in Chapter 1, spatial patterns of the aerosol in Chapter 2, temporal patterns in Chapter 4, aerosol/meteorology relationships in Chapter 5, etc. Because knowledge of aerosol sources is key to planning effective control strategies, this chapter synthesizes the evidence and discusses our overall conclusions regarding aerosol sources. The discussion is organized in two parts -- (1) an overview of particulate sources for the entire RAMS network, and (2) urban versus nonurban patterns including considerations of background concentrations and particulate transport.

7.1 SOURCES OF FINE, COARSE, IP, AND TSP

This section describes the sources of particulate matter in St. Louis, viewing the RAMS network in aggregate. Secondary aerosol sources are distinguished from primary aerosol sources, with the primary aerosol sources further distinguished as to specific source type, e.g. automotove exhaust, fuel oil combustion, iron/steel industry, various source types of crustal material, etc.

7.1.1 Fine Fraction of IP

Annual mean concentrations of FINE range from 16 $\mu g/m^3$ to 28 $\mu g/m^3$ and average 21 $\mu g/m^3$ over the RAMS network. Extending Dzubay's chemcial element balance to the full year of 1976 (see Chapter 1) we find that slightly over 50% of FINE, 11 $\mu g/m^3$ averaged over the network is ammonium sulfate. That sulfate aerosols are the single most important source of FINE is also evidenced by our analysis of spatial patterns, temporal patterns, and aerosol/meteorology relationships; specifically, the features in the data for FINE correspond rather closely to the features for sulfur.

Sulfur concentrations at the rural RAMS sites are approximately 75% of the concentrations at the center-city sites, signifying that the sources of sulfate are of air basin scale if not synoptic scale. Analyses performed in Chapter 3 indicate that the majority of sulfate in the St. Louis area

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apparently results from large scale phenomena and that many of the sources may be exterior to the St. Louis AQCR. That the sulfate phenomenon is also partly air basin in scale (rather than synoptic in scale) is evidenced by the moderate urban/nonurban differences, and by the existence of a weekend-weekday effect (which implies a fairly short response time for the sulfate). The air basin or synoptic scale nature of sulfate implies that it consists predominantly of secondary aerosol produced from SO_2 emissions rather than primary sulfate emissions. Electric power plants are implicated as the major source of sulfate because they contribute about 80% of SO_x emissions in the St. Louis AQCR (EPA, 1978) and 70% of SO_x emissions synoptically (Husar, 1979). Sulfate levels are somewhat elevated in the center-city relative to the countryside due to any of three factors: (1) greater urban SO_2 concentrations which oxidize to form local sulfate; (2) the greater oxidizing potential of the urban atmosphere due to the presence of photochemical smog; and/or (3) the possible importance of primary sulfate emissions from urban sources.

Dzubay's calculations (Chapter 1), the decision-tree analyses (Chapter 5), and the spatial/temporal patterns (Chapters 2 and 4) imply that no single type of aerosol dominates the non-sulfate portion of FINE. Rather, the non-sulfate part of FINE seems to be a mixture of fairly small contributions from numerous (mostly man-made) sources: secondary organic aerosols, secondary nitrates, automotive exhaust particles, particles from stationary source fuel combustion, the fine fraction (i.e. the lower tail of the size distribution) of suspended dust, etc. As discussed in Chapter 5, FINE consists essentially of man-made particulate matter, especially secondary aerosols, especially sulfates.

7.1.2 Coarse Fraction of IP

Annual mean concentrations of COARSE range from 13 $\mu g/m^3$ to 32 $\mu g/m^3$ and average 21 $\mu g/m^3$ among the RAMS sites. Dzubay's chemical element balance (extended to cover the entire year) shows that 83% of COARSE is crustal material: about two-thirds shale type material and one-third limestone type material.

The various patterns that we have observed in the data for COARSE and the crustal elements (Si, Al, and Ca) support Dzubay's conclusion that COARSE is dominated by crustal material, mostly of the shale-type. The analyses in this report have also provided a more refined assessment of the origins of this crustal material. The smooth spatial patterns in the crustal elements (Chapter 2) and the correlation of the crustal elements with dryness (Chapter 5) indicate that ubiquitous area sources of fugitive dust, rather than industrial sources of fly ash, are the dominant contributors to the crustal material. Furthermore, the existence of significant weekend-weekday effects (Chapter 4) and the correlation of the crustal material with low wind speeds (Chapter 5) signify that most of the fugitive dust is man-made rather than wind-blown.

Considering the omnipresence of motor vehicle traffic, one might suspect that road dust constitutes the dominant source of fugitive dust in St. Louis. However, the strong dissimilarities between the spatial/temporal patterns for the crustal elements (or COARSE) and lead suggest that motor vehicles are not the predominant dust source.* Rather, an assortment of dust sources -- paved roads, unpaved roads, quarries, construction, agriculture, soil dust, and certain industries -- seems implicated.

Shale-type and limestone-type crustal material each probably arise from several dust sources in the above list. There is evidence, however, that the limestone-type dust is more closely tied to urban commercial/industrial activity than shale-type dust. The greater urban/rural spatial gradient for calcium (Chapter 2) and the greater weekend effect for calcium (Chapter 5) suggest that crustal limestone stems relatively more from cement manufacturing and construction, while crustal shale stems more from traffic, agriculture, and wind-blown dust.

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*One can invent scenarios whereby traffic could be the predominant source of crustal material, yet crustal material and lead would have incongruous spatial/temporal patterns. For example, the spatial patterns of lead and road dust could diverge because lead emissions are determined by traffic levels and percent gasoline vehicles, while road dust depends on traffic and road conditions (paved versus unpaved, road cleanliness, vehicle speed, etc.); the two independent factors (percent gasoline vehicles and road conditions) could destroy the spatial correlation of road dust and lead. Also, the diurnal patterns of lead and crustal elements may differ because high afternoon mixing heights will disperse the fine lead particles producing the observed afternoon miminum in Pb, while the afternoon effect on coarse dust particles will be to keep more suspended (due to unstable conditions) as well as to disperse them, leading to little net impact on the crustal elements. Despite the potential validity of these arguments, we interpret the data to imply that road dust is not likely the predominant source of the crustal material.

Dzubay's chemical element balance shows that the remaining, non-crustal 17% of COARSE stems from a wide variety of sources (mostly man-made). Our analyses do not reveal very much about this remaining 17% except to reemphasize Dzubay's findings that it is mostly man-made (see decision-tree analyses in Chapter 5). This non-crustal fraction may not be very significant in terms of the total mass of COARSE, but it may nevertheless be important to health and welfare effects because its chemical composition differs from that of crustal material.

7.1.3 Total IP

Annual mean concentrations of IP range from 29 to 60 $\mu g/m^3$ among the RAMS sites and average 42 $\mu g/m^3$. The major sources of IP are the major sources of its two constituents, FINE and COARSE, each of which comprises 50% of IP. The chemical element balance, modified to encompass all of 1976, implies that over three-fourths of IP consists of sulfate (29%) and crustal material (47%). Discussions of the specific sources of sulfate, other parts of FINE, crustal material, and other parts of COARSE are presented in the previous two subsections.

It is noteworthy that nearly half of IP in St. Louis consists of crustal material and that most of the crustal material is apparently dust related. One implication of this result is that going from a TSP air quality standard to an IP air quality standard will not completely eliminate the fugitive dust problems associated with the TSP standard.

7.1.4 TSP

Annual mean concentrations of TSP range from 53 to 96 $\mu g/m^3$ and average 79 $\mu g/m^3$ over the RAMS network. The relationship between particulate air quality and meteorology (Chapter 5) indicates that TSP is more influenced by dust than IP; in fact, dust appears to be the dominant source of TSP. This makes sense because TSP essentially consists of IP plus particles in the 20 to 50 μm size range; and one would expect these larger particles to be composed almost completely of dust.

The spatial pattern of TSP concentrations among the RAMS stations exhibits very smooth spatial gradients, suggesting that the influence of localized sources tends to be small. This reflects the fact that the RAMS stations are generally sited away from strong local sources (fugitive dust or other types). In this sense, the RAMS particulate sites are not representative of existing Hi-Vol sites nationwide, many of which are significantly affected by local sources (especially road dust) because of their proximity to streets, parking lots, and other particulate sources (Record et al., 1976; Pace, 1978; Price et al., 1977).*

7.2 URBAN VERSUS RURAL SOURCES OF IP

In the sense that crustal material and sulfates are the two major components of IP, the origins of IP are generally similar at the five urban sites (103, 105, 106, 108, and 112) and the two rural sites (122 and 124). As evidenced by the urban/rural ratios in crustal elements (see Table 2.2), the relative importance of crustal material is minutely greater at urban sites than rural sites, basically because limestone-type dust is relatively more important at the urban sites. Sulfate, on the other hand, is slightly more significant at nonurban locations; ammonium sulfate comprises 27% of urban IP but 33% of rural IP. Essentially because of the sulfate pattern, FINE makes up 49% of urban IP but 53% of rural IP.

There are, however, some notable differences in the sources of urban and rural particulate matter. The spatial patterns of lead, vanadium, titanium, and iron imply that primary aerosols from auto exhaust, fuel combustion, and industrial sources are typically a factor of three to five times greater at urban sites than rural sites. Although the particulate matter from these sources is not a major portion of IP mass, it may be of concern because of its special chemical composition (i.e. it may be more toxic than crustal material). Also, as evidenced by air quality/meteorology relationships (Chapter 5) and weekend-weekday effects (Chapter 4) for the crustal elements, the specific sources of crustal material appear to differ significantly from urban to rural sites. Urban dust sources (road dust, construction, cement manufacturing, industrial dust, etc.) are relatively more important in the city, while agriculture and wind-blown dust are relatively more important in the countryside. Of course, we generally expect natural sources to contribute a larger fraction of the particulate matter at rural locations.

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*In a study of 154 Hi-Vol sites in 14 cities, Record et al. (1976) found that 45% of the sites had some degree of local impact, 22% had major local impacts, and 8% had "undue" local impacts.

The analysis of background concentrations and particulate transport (Chapter 3) indicated that transport of aerosols from the urban area did not have a major effect on the rural sites. On an annual mean basis, transport from the urban area appears to contribute less than 5 $\mu g/m^3$ of aerosol (basically FINE) to the rural sites. Considerable uncertainty existed in our estimates of the transport effect as well as in our estimates of background concentrations. Annual mean background concentrations (representing contributions from natural sources and from man-made sources exterior to the St. Louis AQCR) appear to be approximately 35 $\mu g/m^3$ for TSP and 20 $\mu g/m^3$ for IP.

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